Workshop on correlated electron systems – novel developments
Fine Theoretical physical Institute, Minneapolis – May 16, 2018

Hund's correlated Metals

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EDUCATION SCIENCE INNOVATION

Hund's metals

LETTERS

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nature materials

Kinetic frustration and the nature of the magnetic and paramagnetic states in iron pnictides and iron chalcogenides

Z. P. Yin[1,2]*, K. Haule[1] and G. Kotliar[1]

The iron pnictide and chalcogenide compounds are a subject of intensive investigations owing to their surprisingly high temperature superconductivity[1]. They all share the same basic building blocks, but there is significant variation in their physical properties, such as magnetic ordered moments, effective masses, superconducting gaps and transition temperature (T_c). Many theoretical techniques have been applied to individual compounds but no consistent description of the microscopic origin of these variations is available[2]. Here we carry out a comparative theoretical study of a large number of iron-based compounds in both their magnetic and paramagnetic states. Taking into account correlation effects and realistic band structures, we describe well the trends in all of the physical properties such as the ordered moments, effective masses and Fermi surfaces across all families of iron compounds, and find them to be in good agreement with experiments. We trace variation in physical properties to variations in the key structural parameters, rather than changes in the screening of the Coulomb interactions. Our results also provide a natural explanation of the strongly Fermi-surface-dependent superconducting gaps observed in experiments[3].

The iron pnictides/chalcogenides are Hund's metals. In these systems the Coulomb interaction among the electrons is not strong enough to fully localize them, but it significantly slows them down, such that low-energy emerging quasiparticles have a substantially enhanced mass[4]. This enhanced mass emerges not because of the Hubbard interaction U, but because of the Hund's rule interactions that tend to align electrons with the same spin but different orbital quantum numbers when they find themselves on the same iron atom[4].

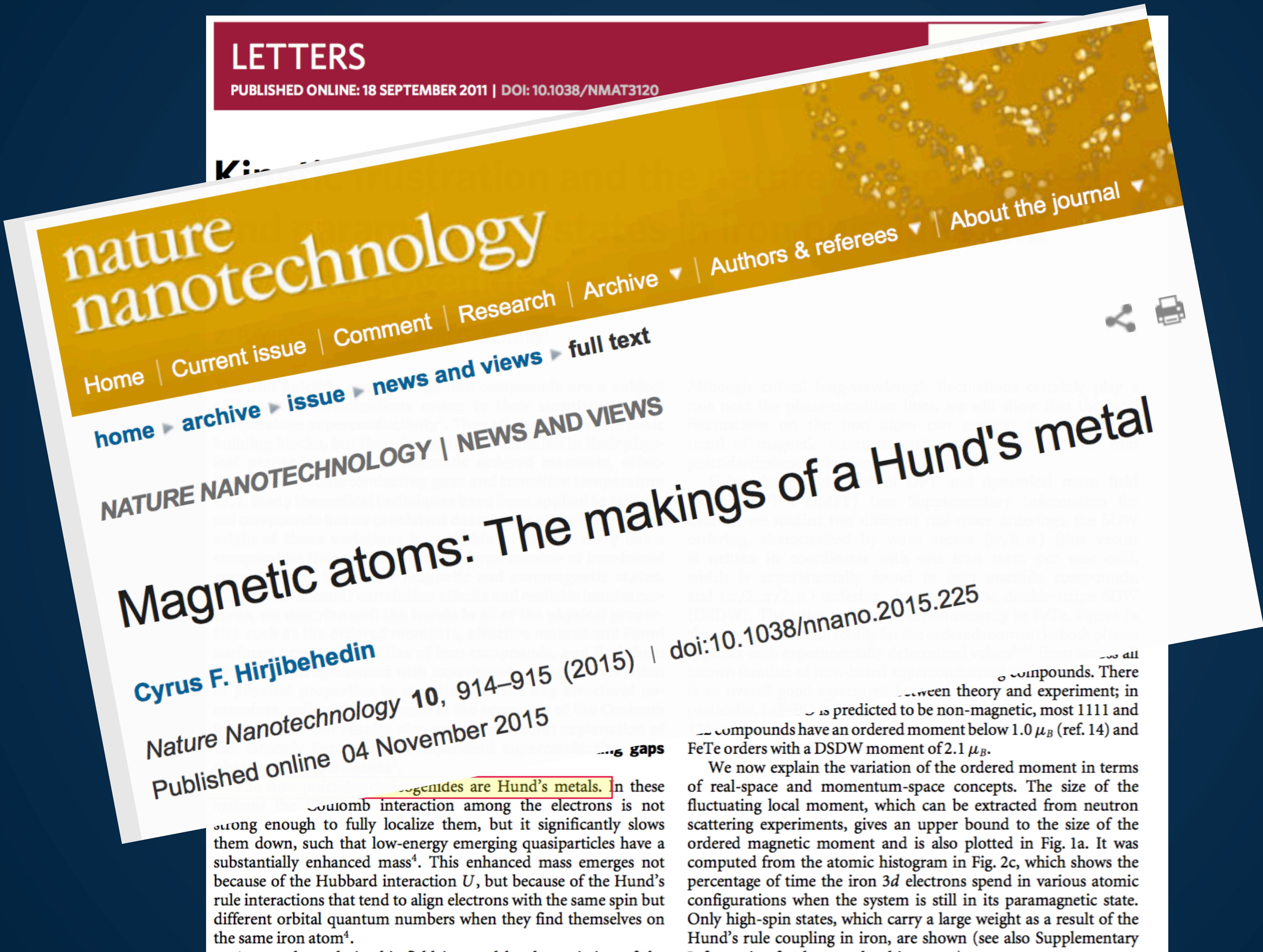
Although critical long-wavelength fluctuations certainly play a role near the phase-transition lines, we will show that the local fluctuations on the iron atom can account for the correct trend of magnetic moments and correlation strength in iron pnictide/chalcogenide layered compounds.

Using the combination of DFT and dynamical mean field theory (DFT + DMFT) (see Supplementary Information for details), we studied two different real-space orderings, the SDW ordering, characterized by wave vector $(\pi, 0, \pi)$ (this vector is written in coordinates with one iron atom per unit cell), which is experimentally found in iron arsenide compounds, and $(\pi/2, \pi/2, \pi)$ ordering, denoted by the double-stripe SDW (DSDW). The latter was found experimentally in FeTe. Figure 1a shows our theoretical results for the ordered moment in both phases together with experimentally determined values[6–13] from across all known families of iron-based superconducting compounds. There is an overall good agreement between theory and experiment; in particular, LaFePO is predicted to be non-magnetic, most 1111 and 122 compounds have an ordered moment below 1.0 μ_B (ref. 14) and FeTe orders with a DSDW moment of 2.1 μ_B.

We now explain the variation of the ordered moment in terms of real-space and momentum-space concepts. The size of the fluctuating local moment, which can be extracted from neutron scattering experiments, gives an upper bound to the size of the ordered magnetic moment and is also plotted in Fig. 1a. It was computed from the atomic histogram in Fig. 2c, which shows the percentage of time the iron 3d electrons spend in various atomic configurations when the system is still in its paramagnetic state. Only high-spin states, which carry a large weight as a result of the Hund's rule coupling in iron, are shown (see also Supplementary

Yin et al, Nat Mat 10, 932 (2011)

Hund's metals



Yin et al, Nat Mat 10, 932 (2011)

Hund's metals

Yin et al, Nat Mat 10, 932 (2011)

Hund's metals

Yin et al, Nat Mat 10, 932 (2011)

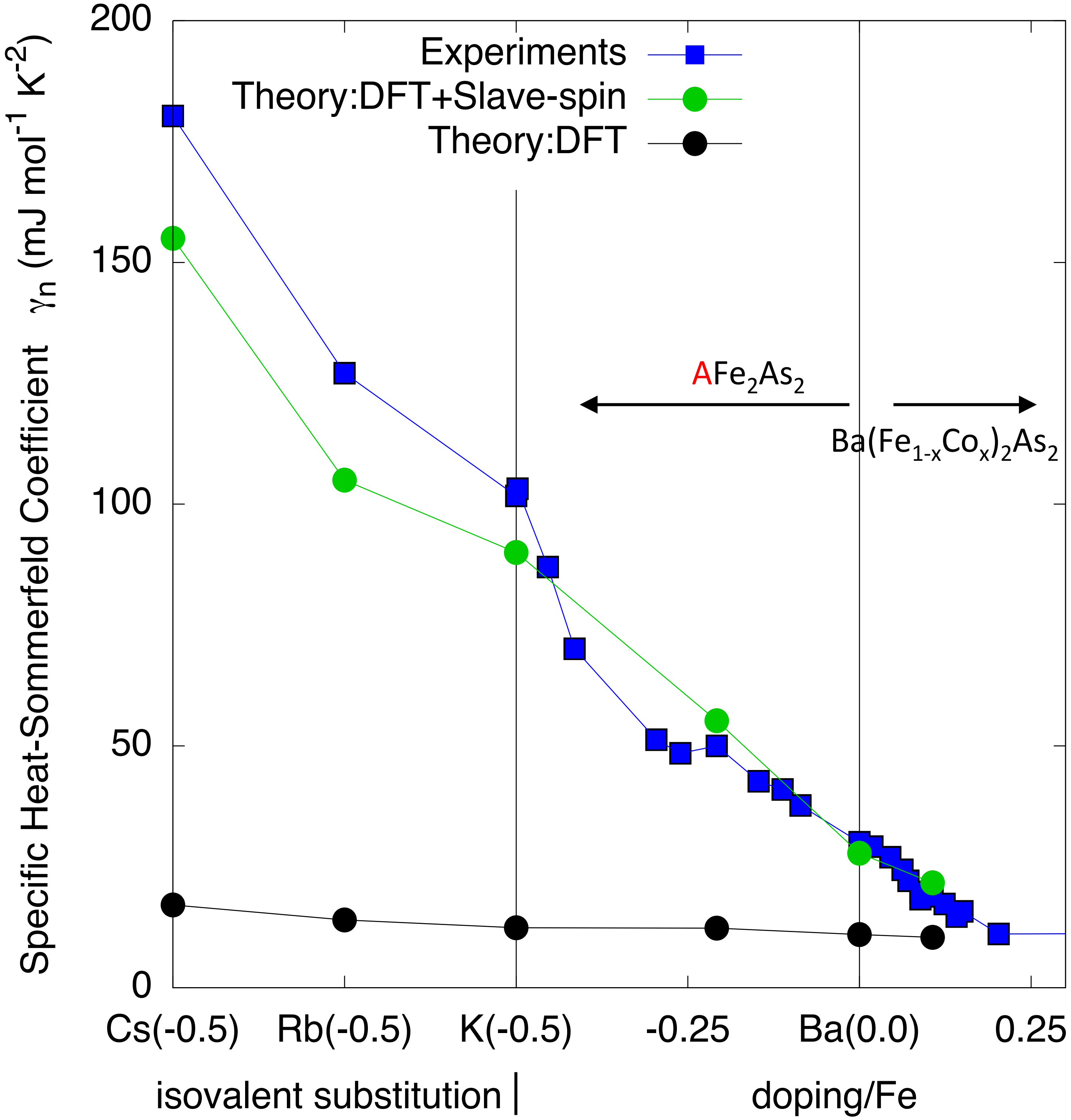
Which correlations for high-Tc superconductivity?

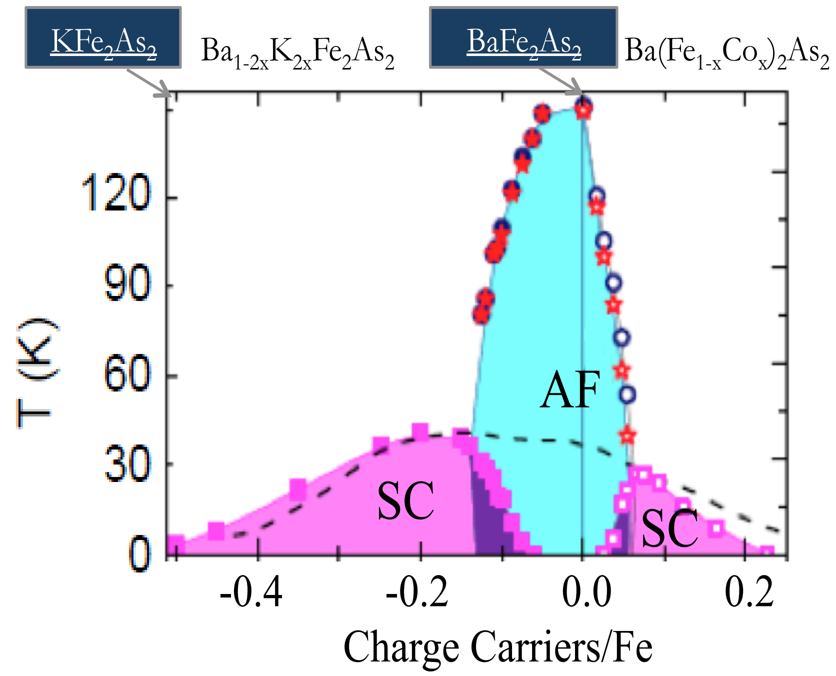
<u>correlations:</u> weak intermediate strong selective ?

Importance in Fermi-liquid pictures:

- renormalization of the quasiparticle bandstructures
- renormalization of quasiparticle interactions?

Fe-superconductors: specific heat



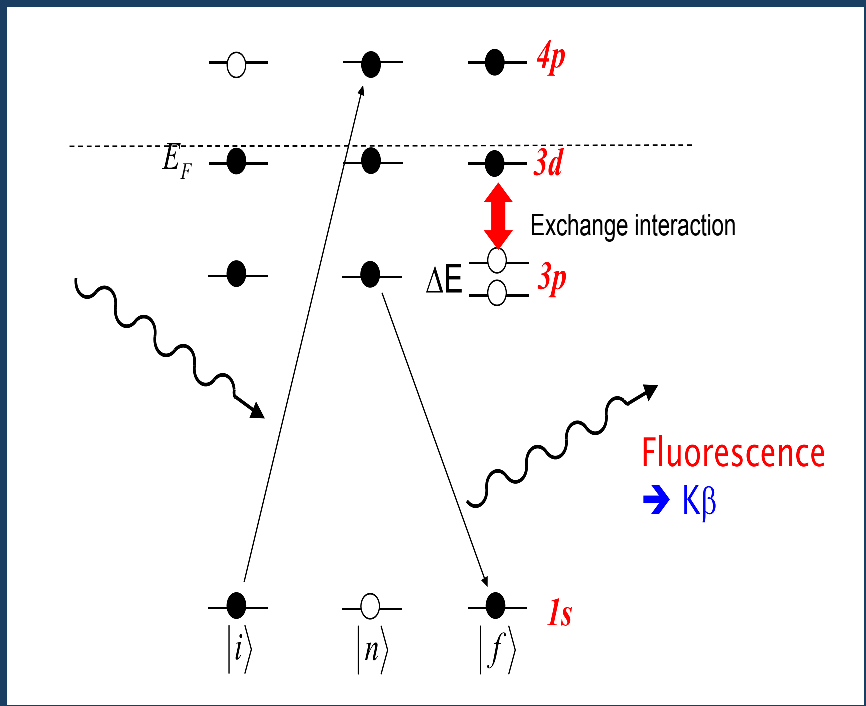


Theory (DFT+Slave-spin):
- the same interaction parameters (U=2.7eV, J/U=0.25) capture the whole material trend
- DFT results are completely off: strong correlations

Experiments: C. Meingast's group in Karlsruhe. F. Hardy,…, LdM et al. PRB 94, 205113 (2016)

Fe-superconductors: local moments

X-ray Emission Spectroscopy
(K-β/β' line shift)



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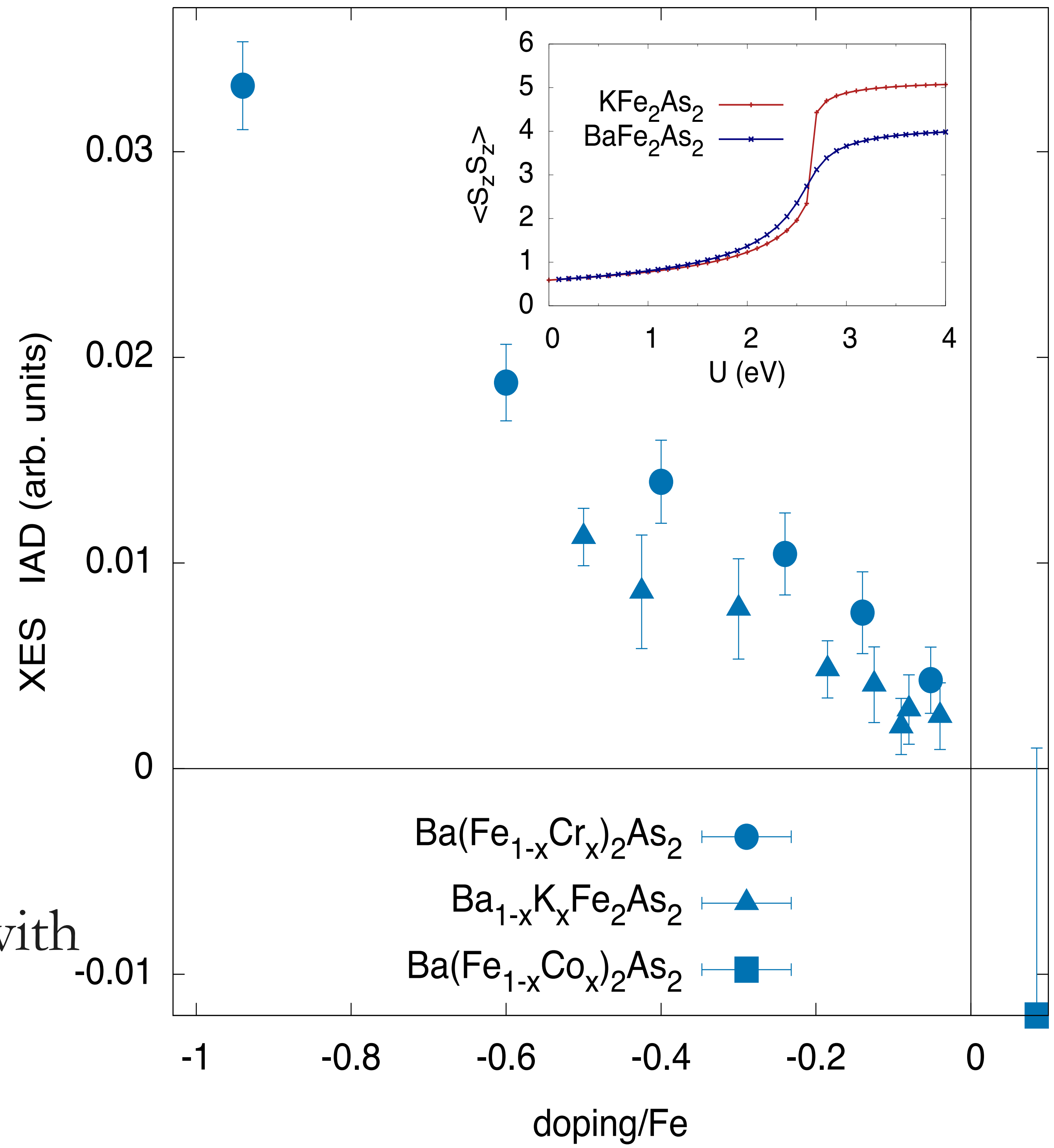


Instantaneous local moments building with hole-doping in 122 Fe-pnictides!

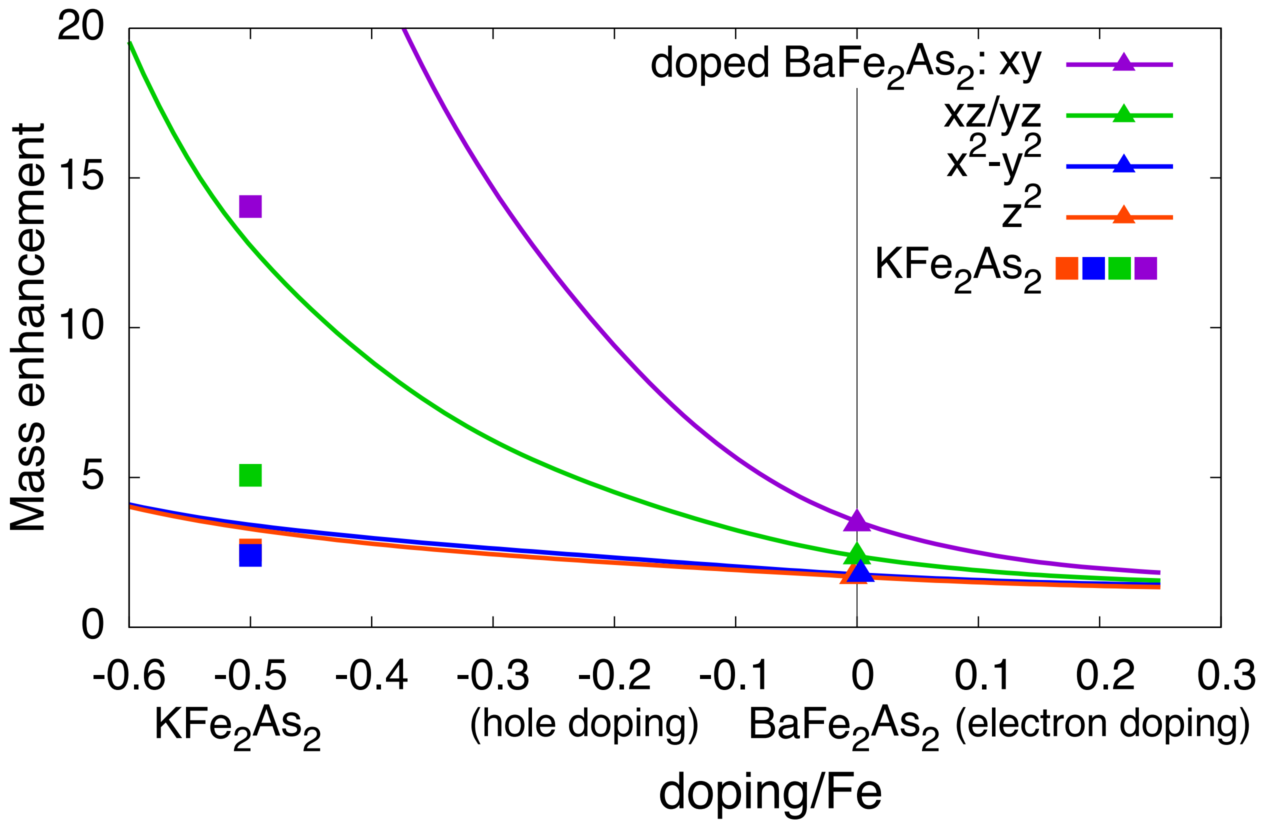
P. Glatzel group @ ESRF (ID26)

S. Lafuerza, …, LdM et al., PRB 96, 045133 (2017)

see also Pelliciari et al. Sci. Rep. 7, 8003 (2017)

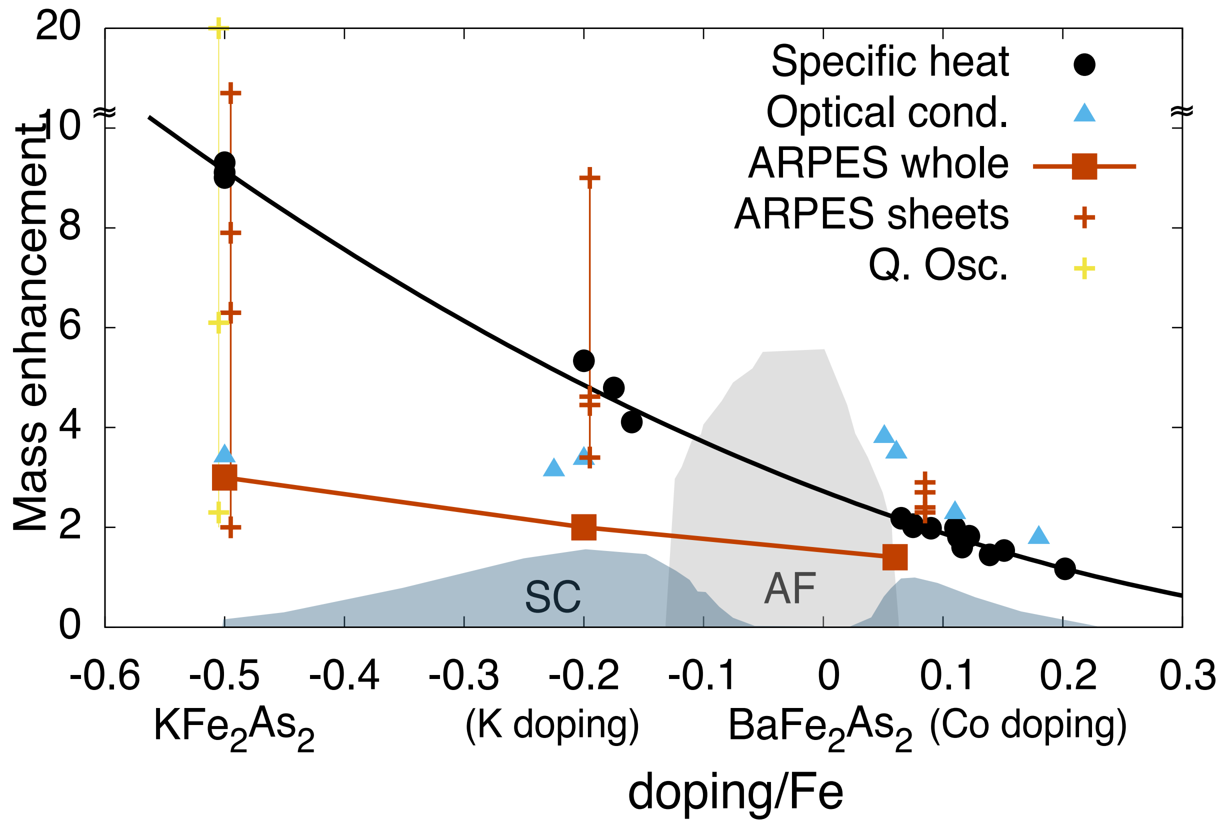
Fe-superconductors: orbital-selectivity

<u>Theory</u> (LDA+Slave-spins)



<u>Experimental data</u>

(high-T tetragonal phase)



LdM, Giovannetti, Capone, PRL 2014 "Selective Mott Physics as a Key to Iron Superconductors"

Selective correlation strength:

strongly *and* weakly correlated electrons coexisting

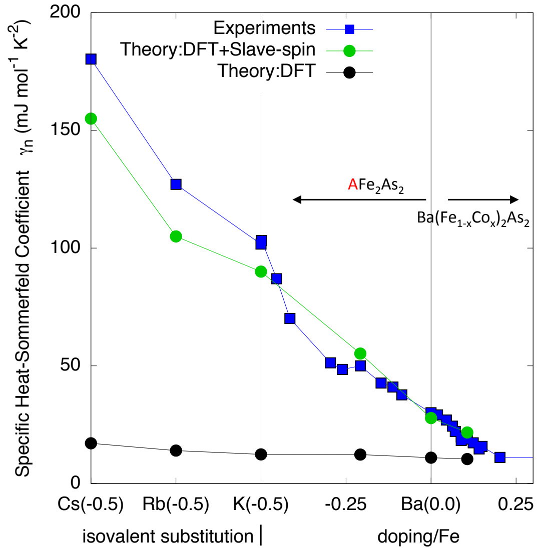
LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015

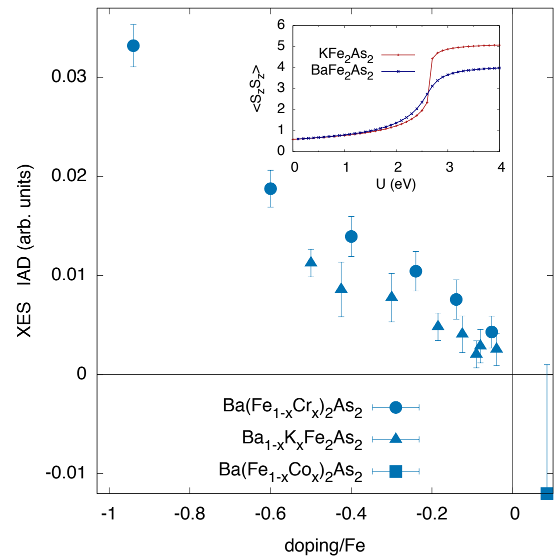
Original proposal (selectivity in FeSC, <u>xy most correlated</u>): LdM et al. PRL2009, J Superc. Nov. Mag. 2009

Hund’s metals

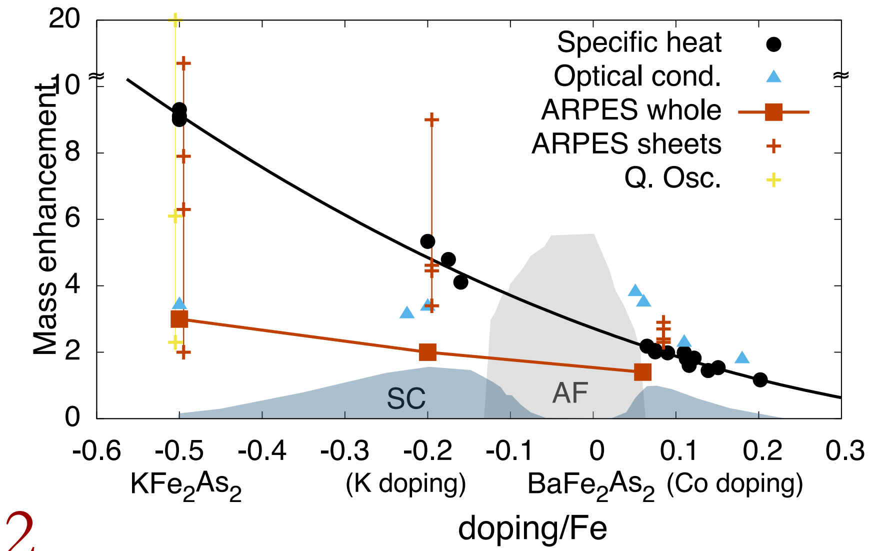
3 main features:

- enhanced electron correlations and masses
- high local spin configurations dominating the paramagnetic fluctuations
- orbital-selectivity of the electron correlation strength





which are due to the proximity to a Hund’s-favored <u>Mott insulating state for half-filled conduction bands</u> (1 hole/Fe doping)

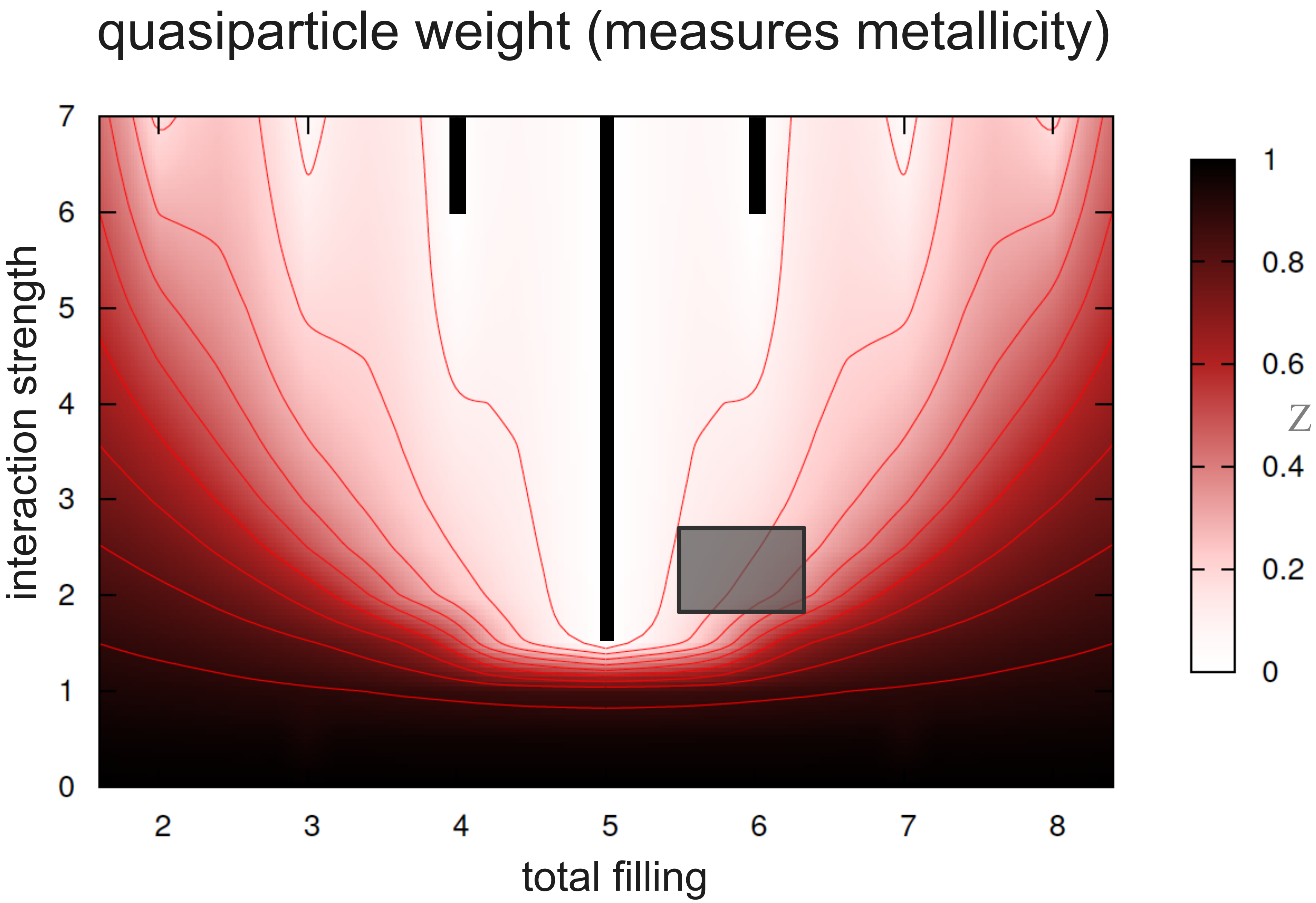


LdM, *Hund’s metals explained*, Juelich lecture, ArXiv:1707.03282

Hund's metal and half-filled Mott insulator

5-orbital Hubbard model

J/U=0.20



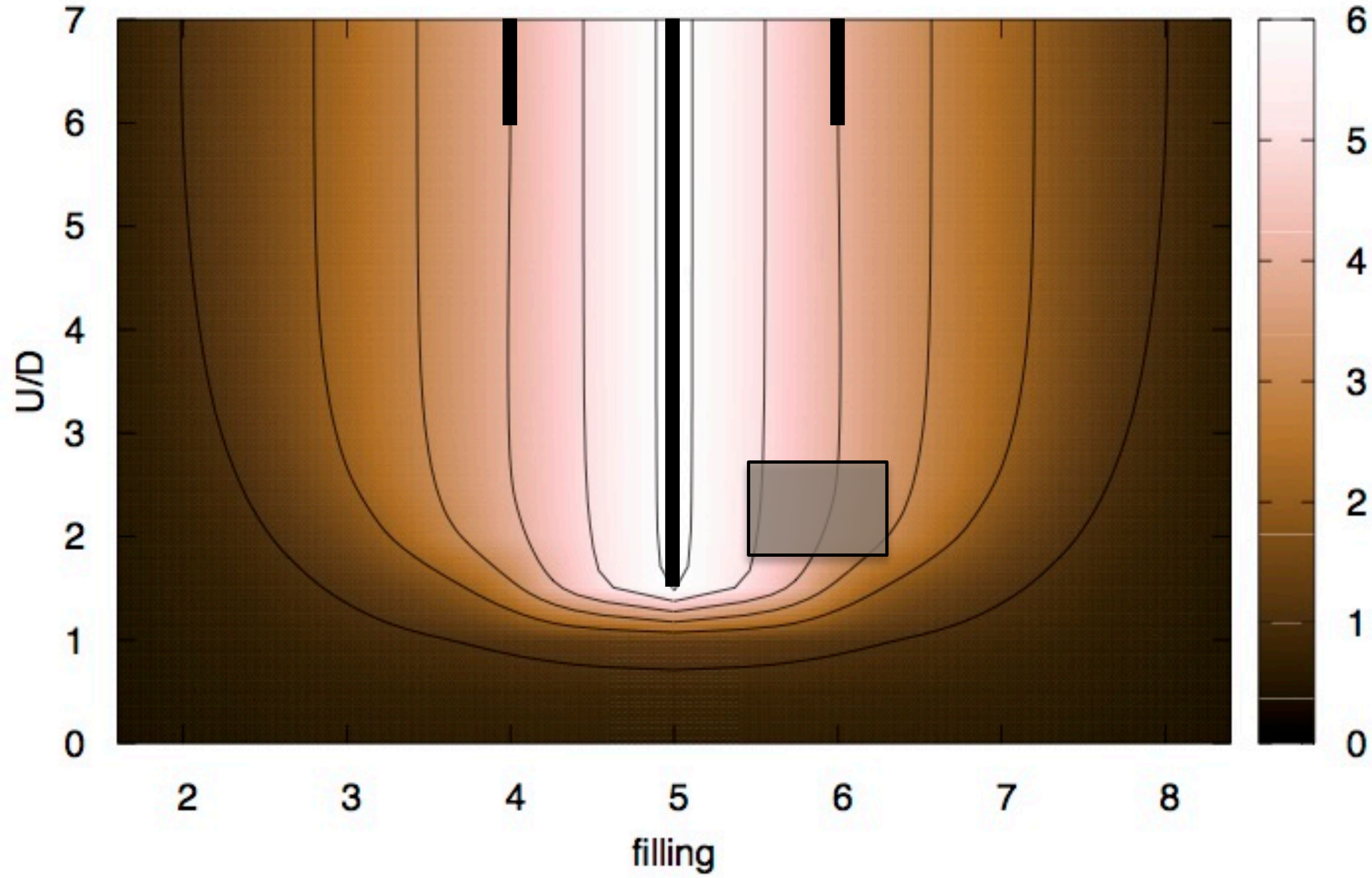
A half-filled Mott insulator dominates the phase diagram

LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015

High fluctuating magnetic moment

5-orbital Hubbard model

J/U=0.20



A metal in which high-spin configurations prevail

"Hund's metal"

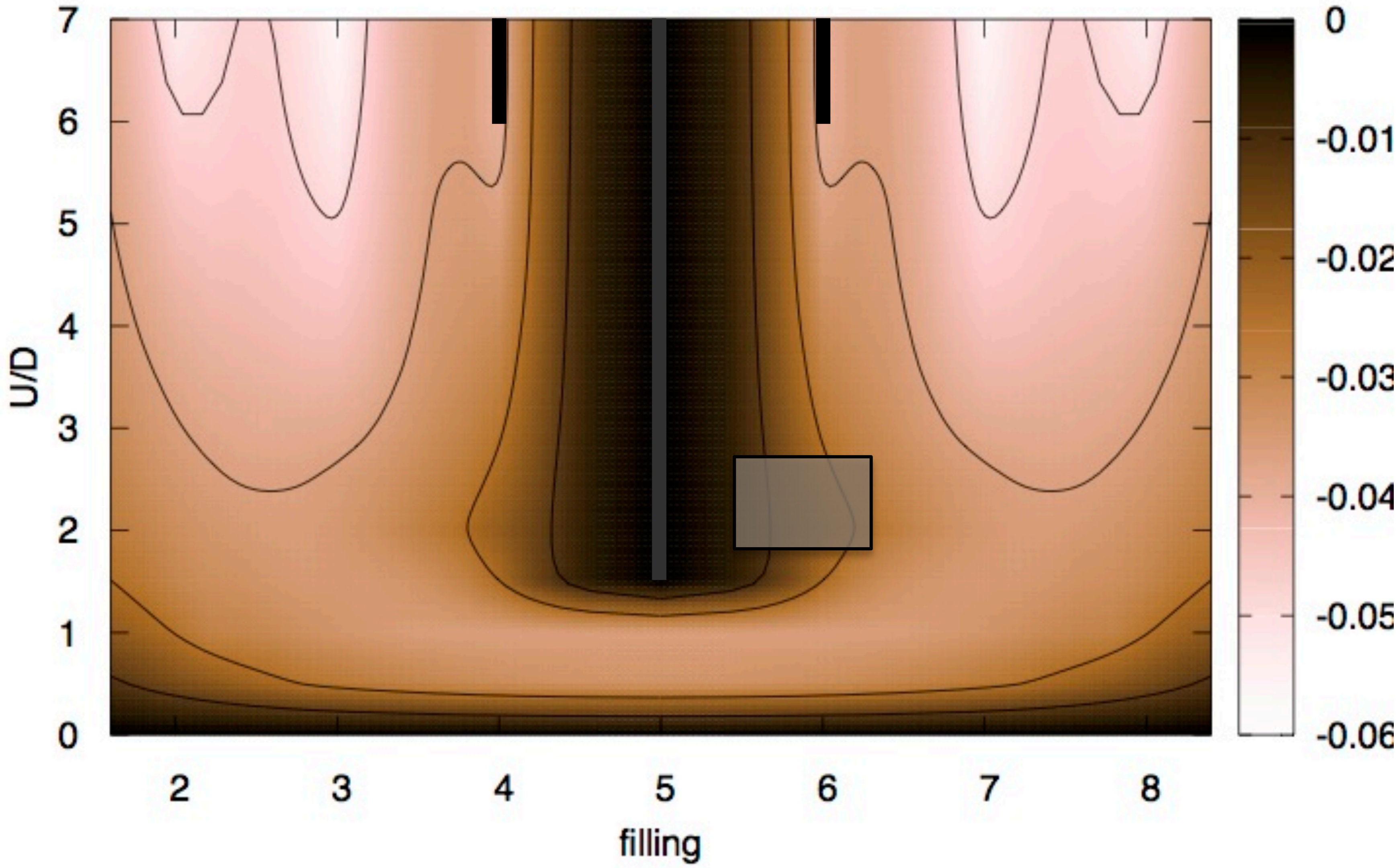
LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015

inter-orbital charge correlations

5-orbital Hubbard model

J/U=0.20

inter-orbital charge correlations $<(n_a-<n_a>)\,(n_b-<n_b>)>$



charge fluctuations in different orbitals become uncorrelated near the half-filled Mott insulator

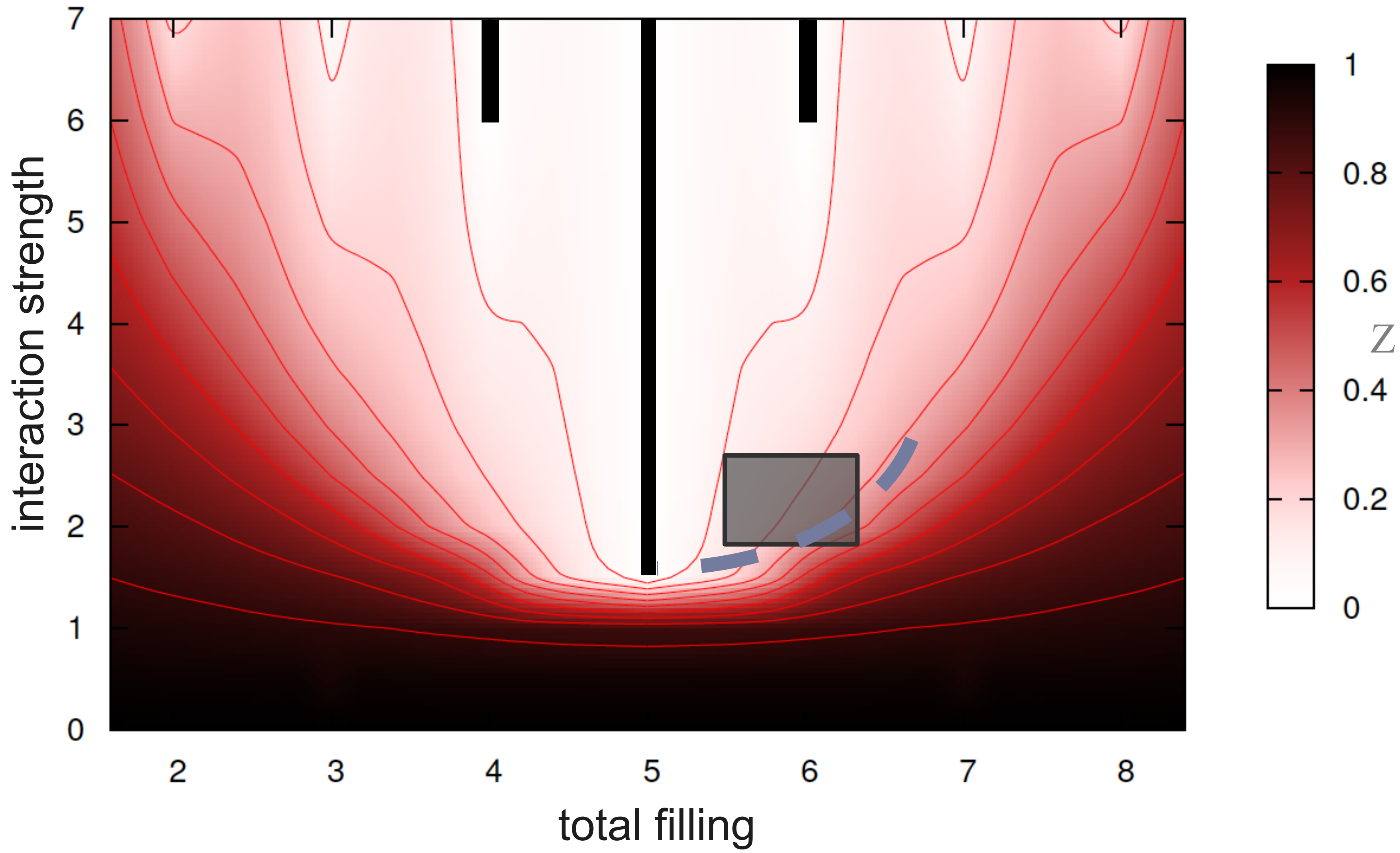
LdM, *Weak AND strong correlations in Fe-SC*, in "Iron-based Superconductivity", Springer book 2015
see also Fanfarillo and Bascones, PRB 92 075136 (2015)

Hund's metal and half-filled Mott insulator

5-orbital Hubbard model

J/U=0.20

quasiparticle weight (measures metallicity)



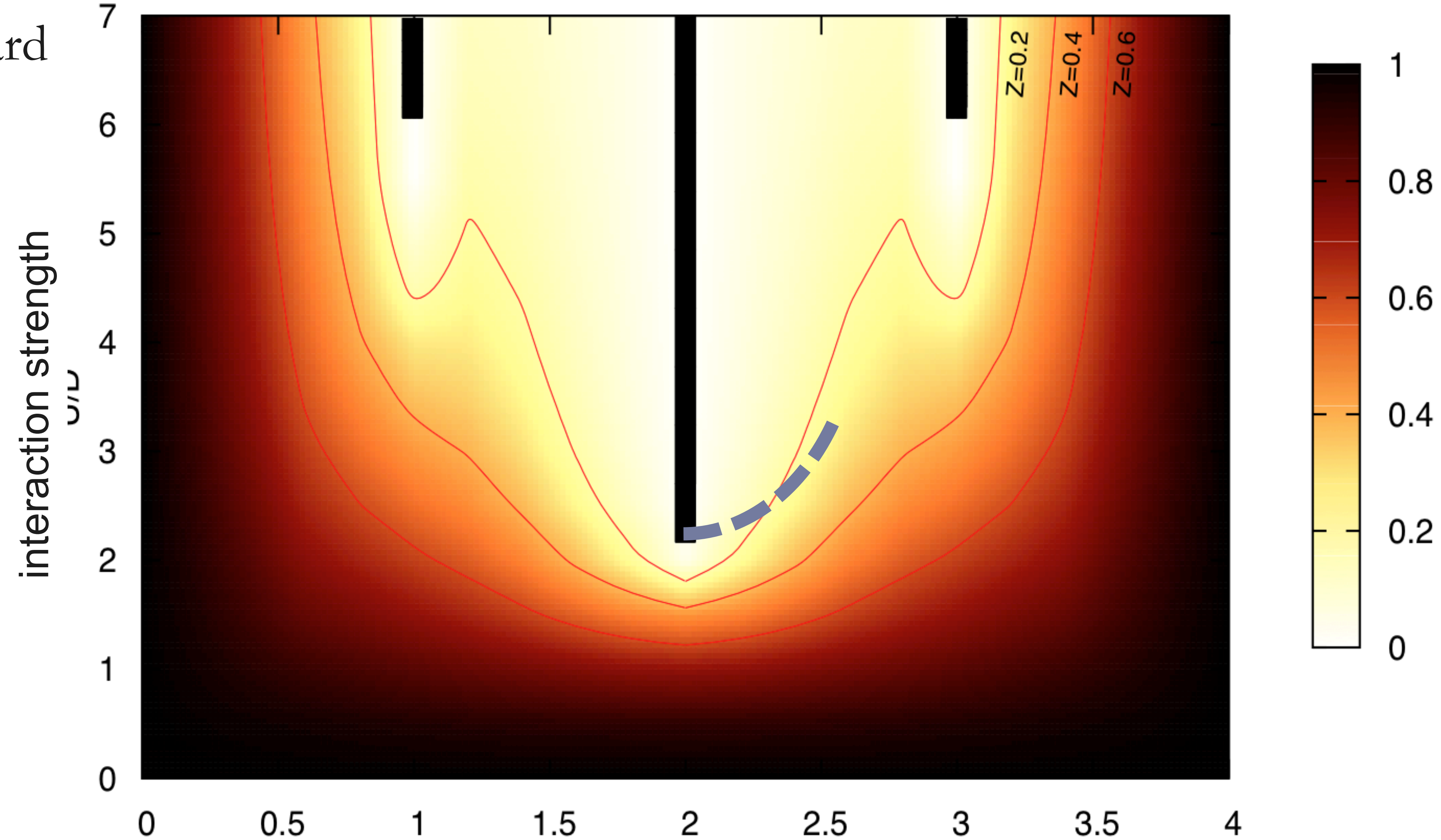
The cross-over departs from the Mott transition at half-filling

frontier first traced in: Ishida and Liebsch, PRB 81, 054513 (2010),
see also Fanfarillo and Bascones , PRB 92 075136 (2015), Misawa and Imada, PRL 108, 177004 (2012)

Hund's metal and half-filled Mott insulator

2-orbital Hubbard model

quasiparticle weight (measures metallicity)

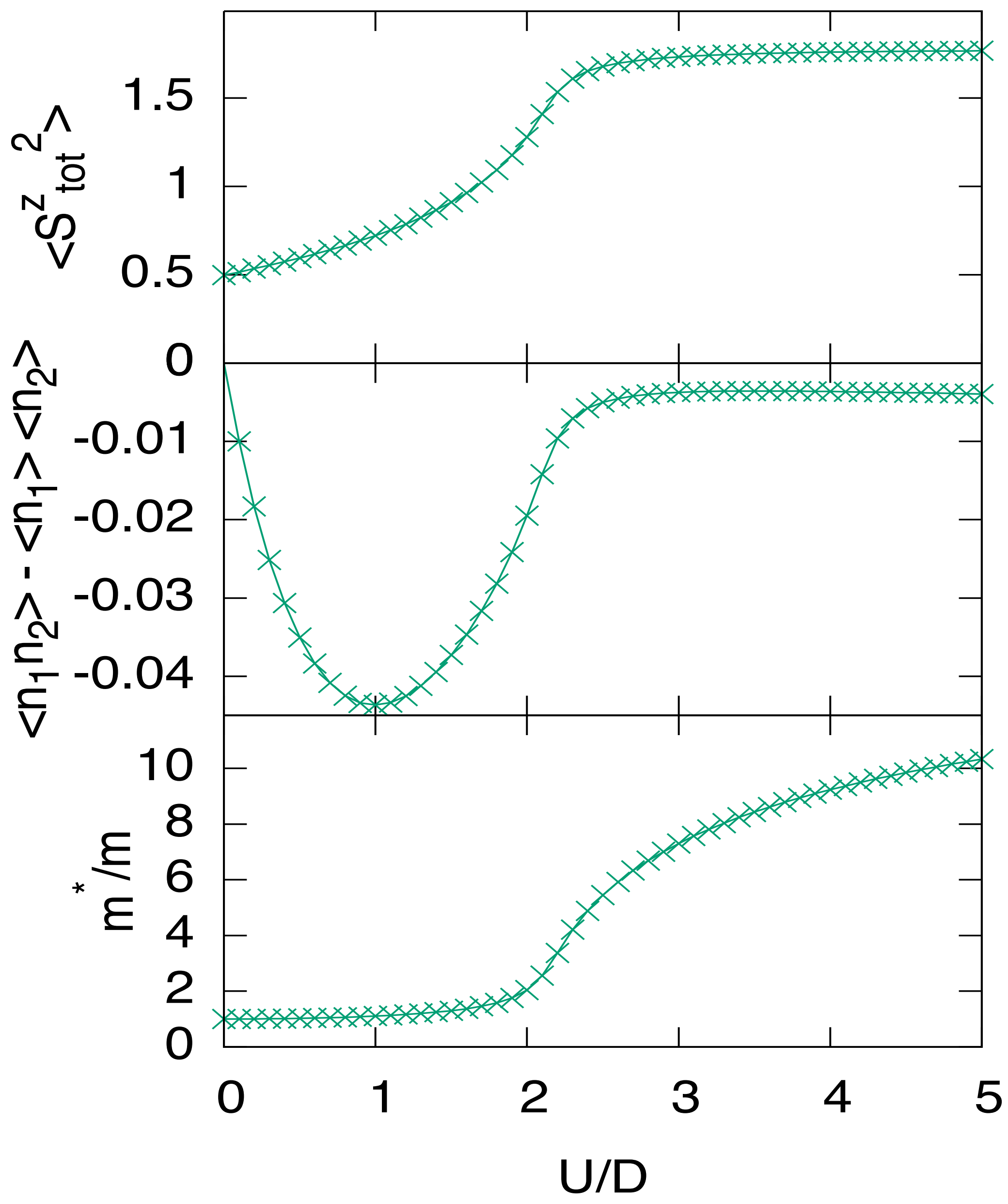


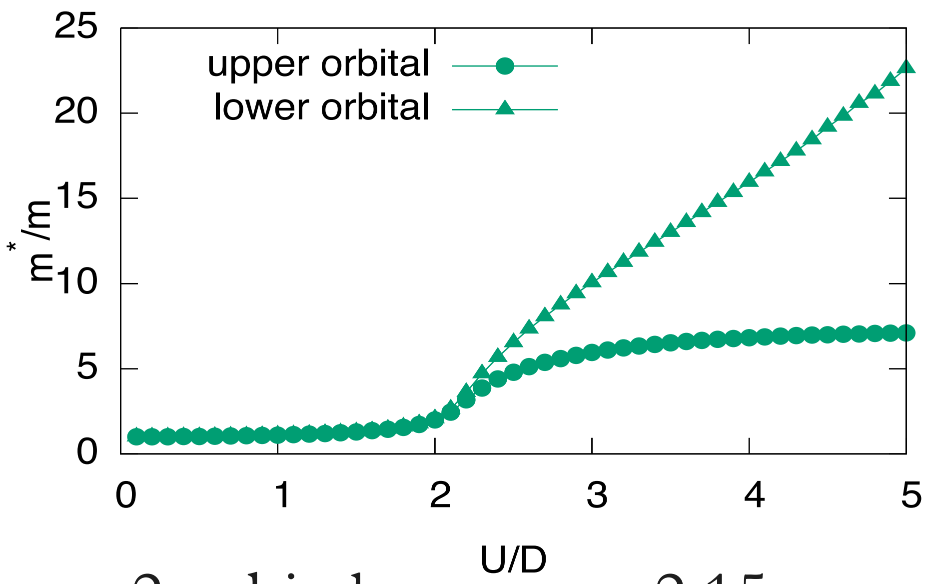
2-orbital Hubbard analogous (and 3-orbital, etc.)

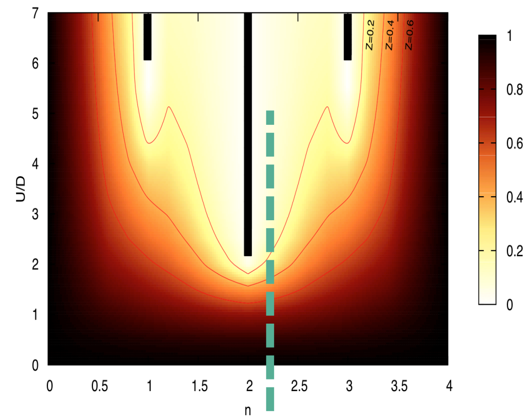
LdM, Mravlje, Georges, PRL 2011, "Janus-faced influence of Hund's rule coupling"

Hund's metal frontier: 2-orbital Hubbard model

2-orbital model: **n=2.15**



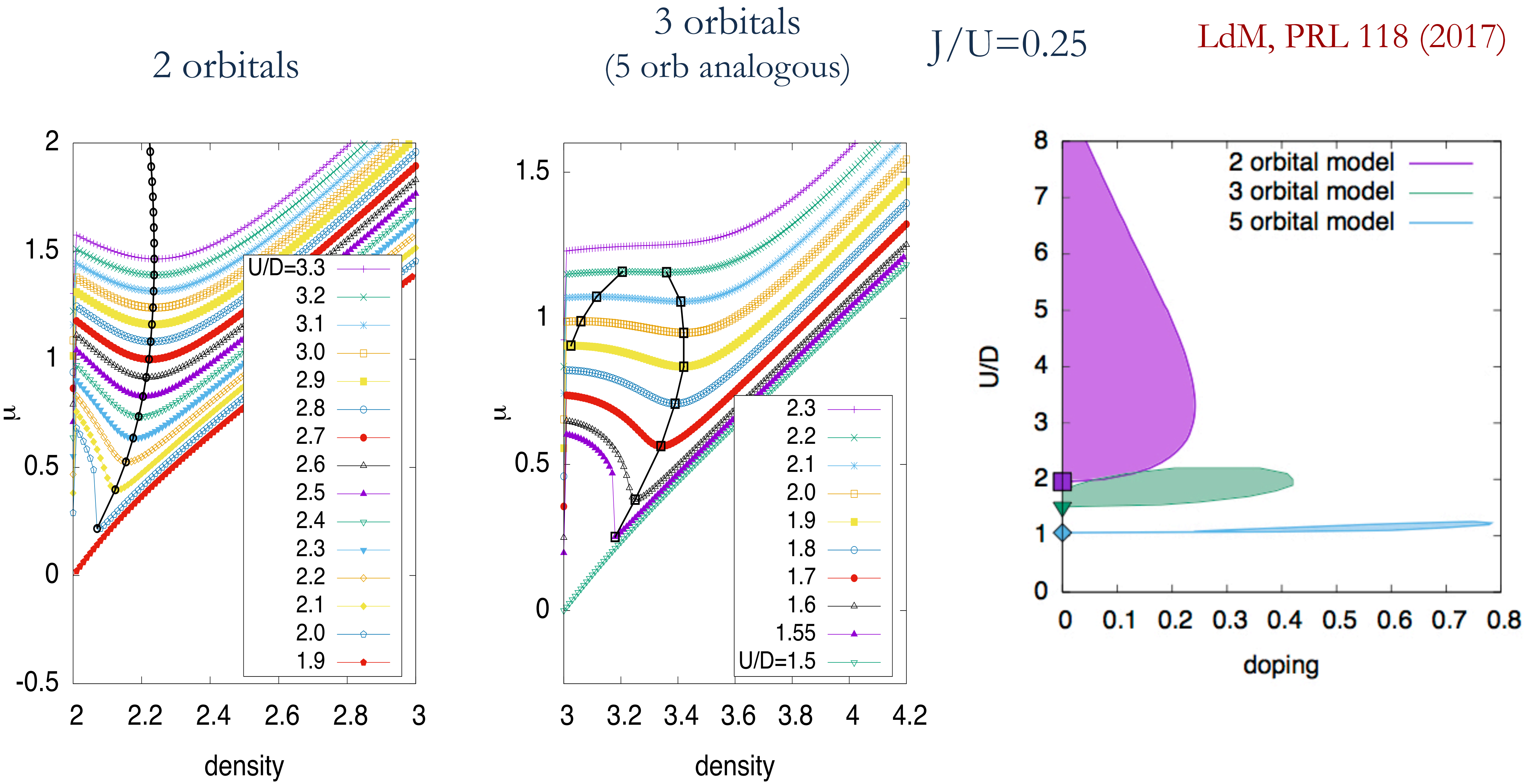




2-orbital case n_{tot}=2.15, with a small crystal-field splitting ε_1-ε_2=0.05D

<u>Hund's phenomenology analogous</u> to the 5-orbital (and 3-orbital) case and to the realistic simulations for the Fe-superconductors: <u>**generic**</u>

Compressibility in 2/3/5-orbital Hubbard model

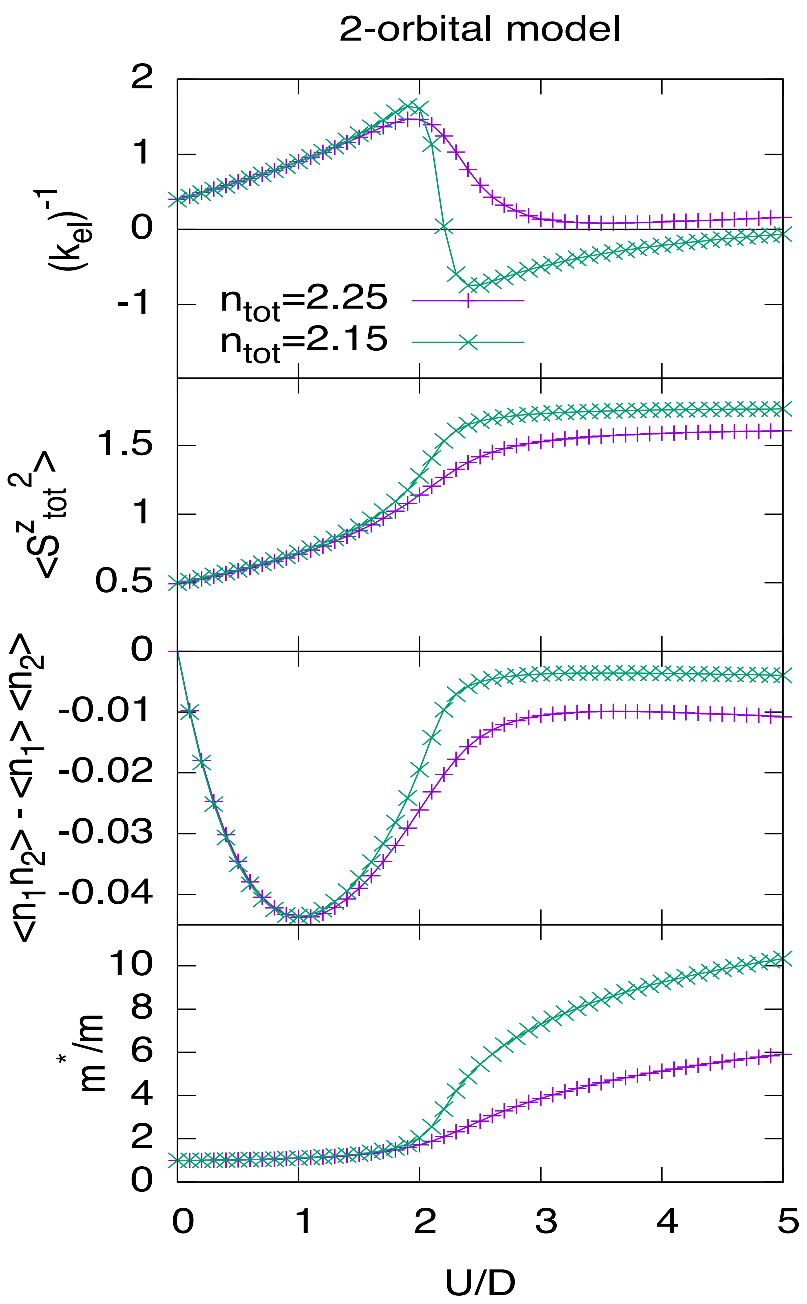


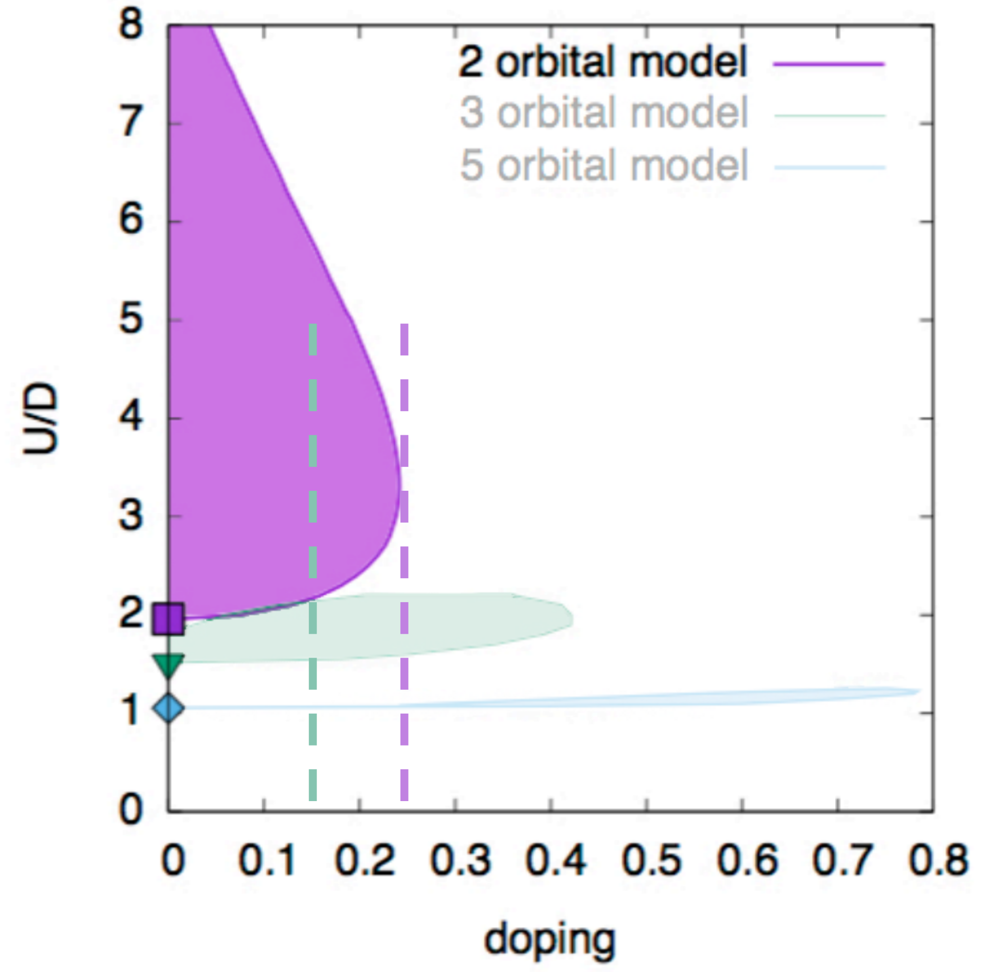
$$\chi = \frac{dn}{d\mu}$$

Divergence of the compressibility on a cross-over line <u>departing from the Mott transition at half filling</u>

Hund's metal frontier and enhanced compressibility

LdM, PRL 118 (2017)

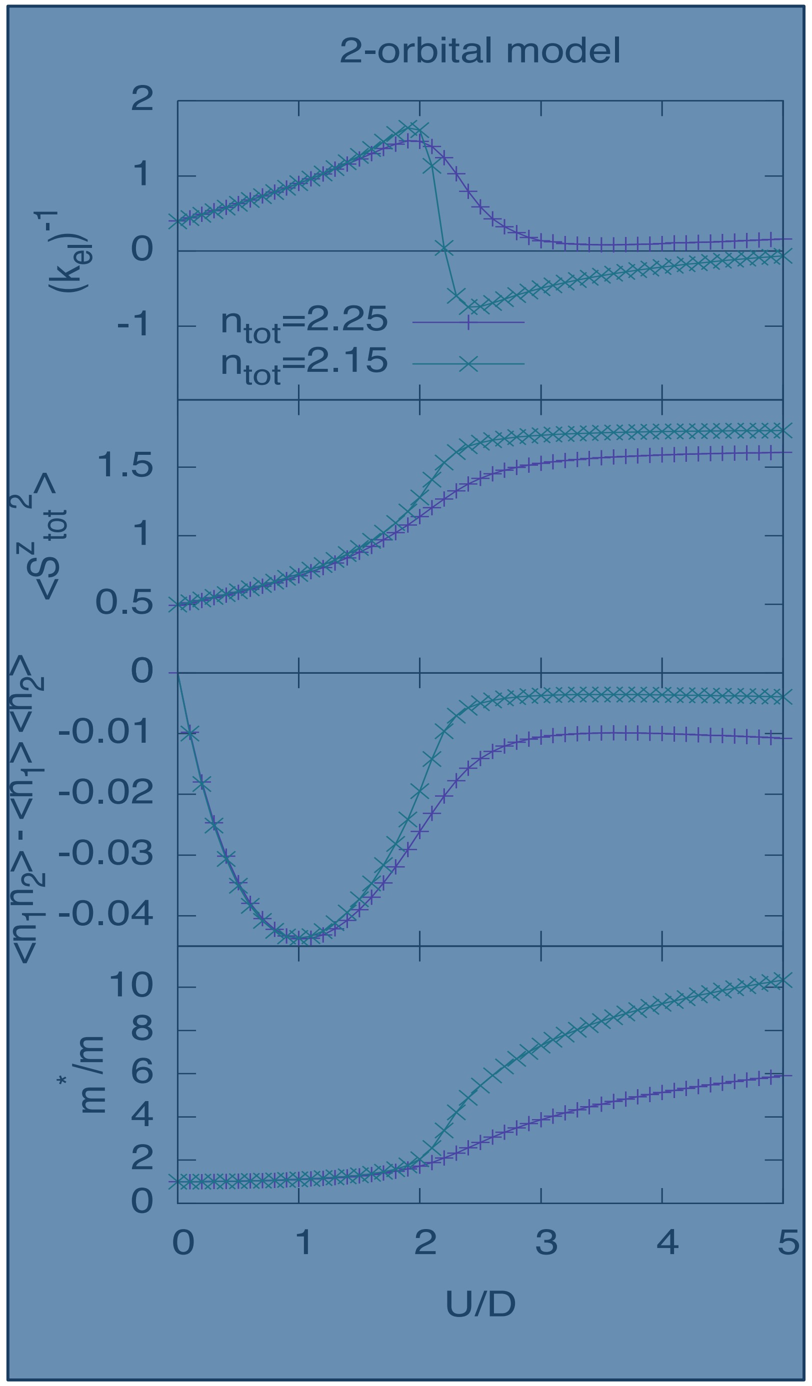


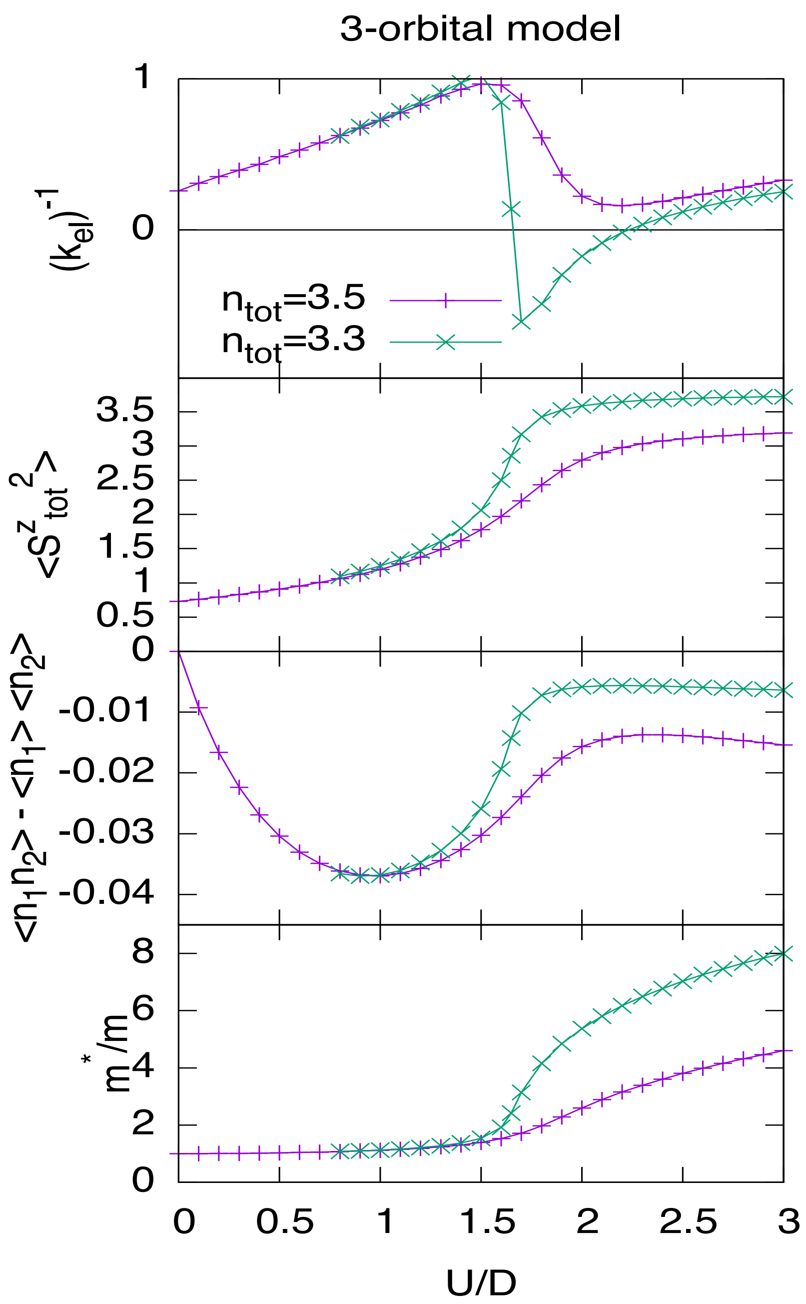


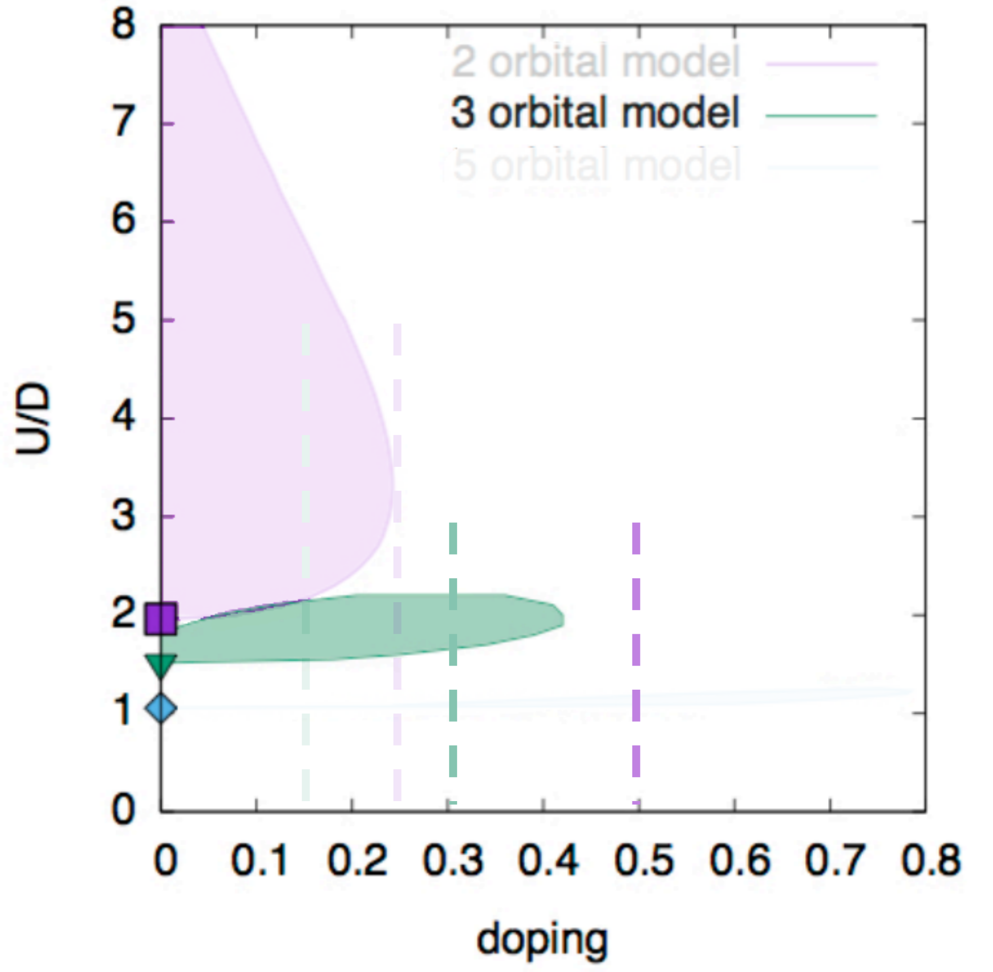
The enhanced/divergent compressibility always occurs <u>near (just inside) the Hund's metal frontier</u>

Hund's metal frontier and enhanced compressibility

LdM, PRL 118 (2017)



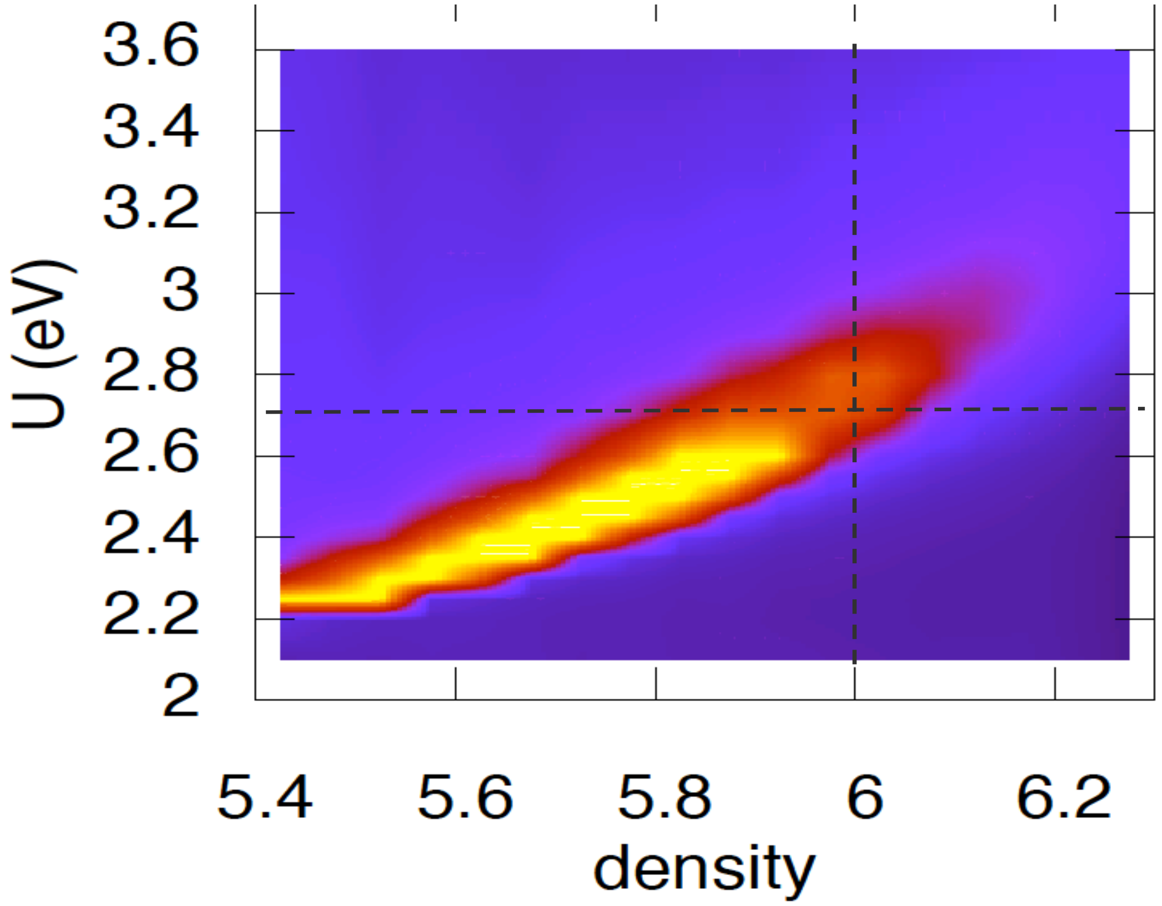




The enhanced/divergent compressibility always occurs <u>near (just inside) the Hund's metal frontier</u>

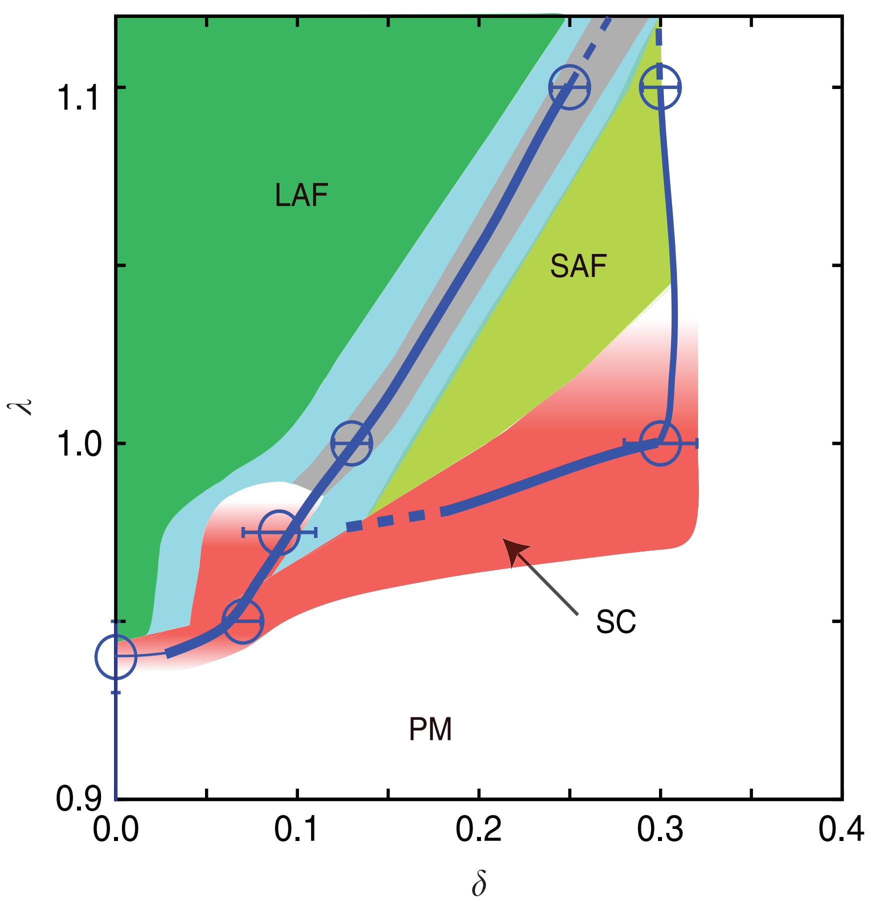
Enhanced compressibility in $BaFe_2As_2$

$BaFe_2As_2$



LdM, PRL 118, 167003 (2017)

LaFeAsO



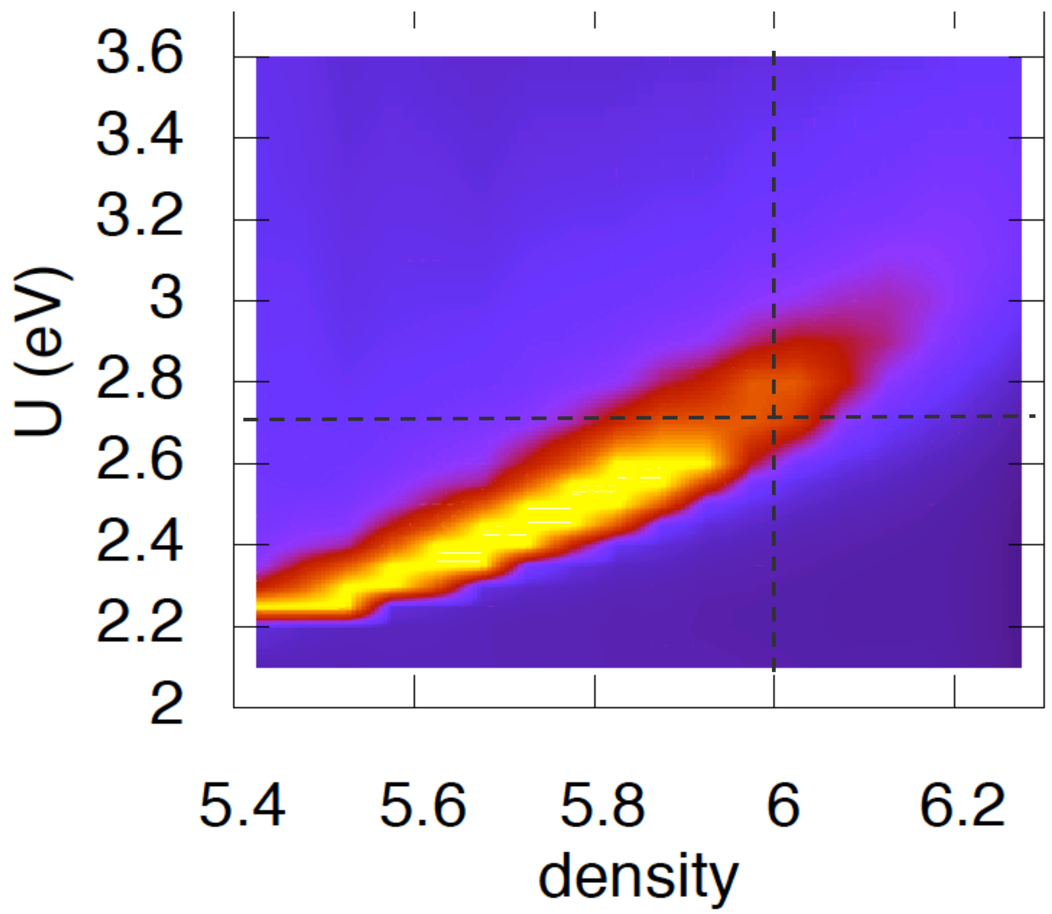
(VQMC)

Misawa and Imada, Nat. Comm 5, 5738 (2014)

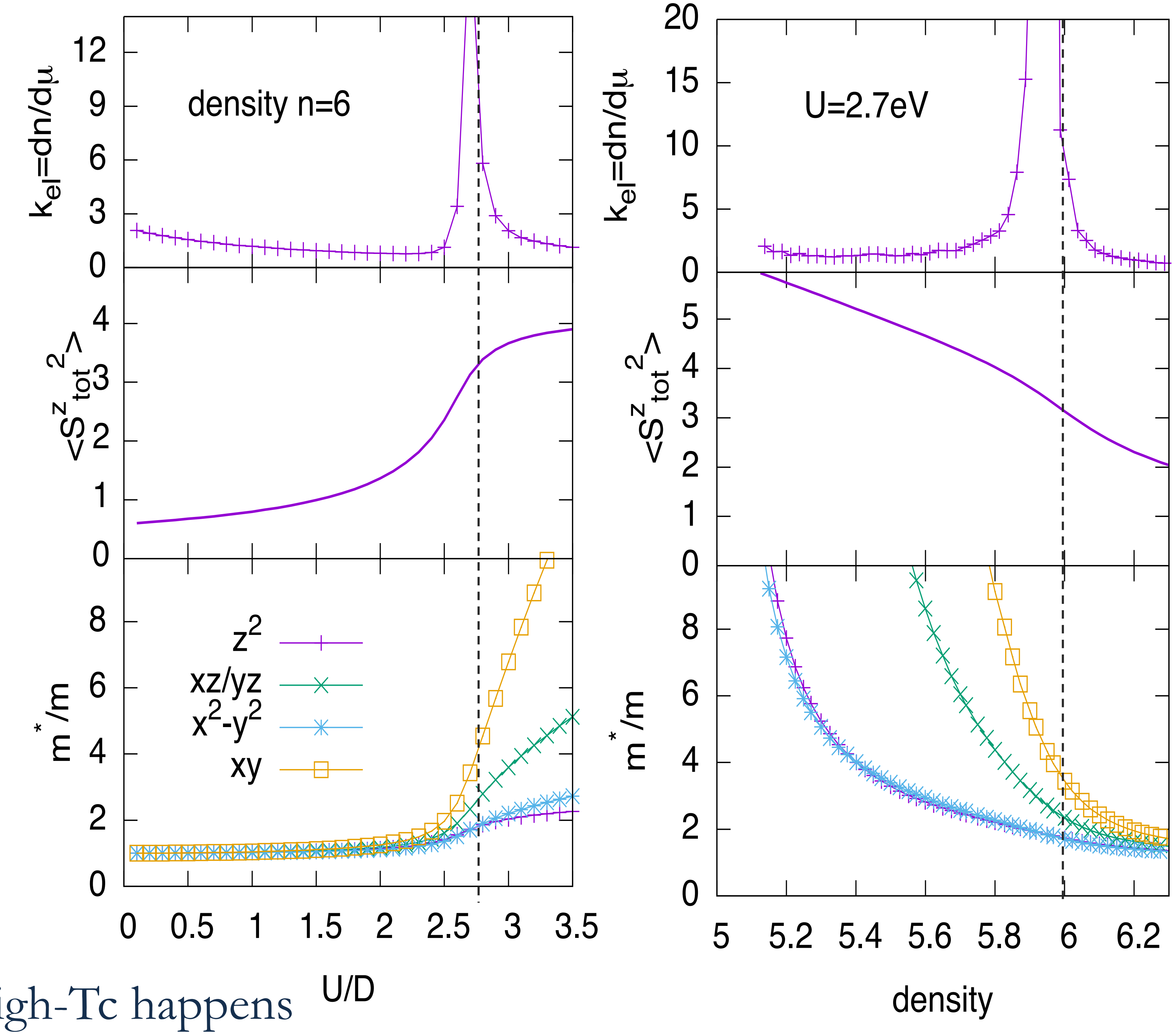
In slave-spin mean-field it is simply a Fermi-liquid instability

- independent of any symmetry breaking
- caused by Hund's coupling
- universal feature of Hund's metals

Enhanced compressibility in $BaFe_2As_2$



LdM, PRL 118, 167003 (2017)



compressibility enhanced:

- in the doping zone where high-Tc happens
- at the entrance of the Hund's metal zone

Enhanced compressibility and superconductivity

In an isotropic Fermi liquid:

- $$\chi = \frac{\chi_0/Z}{1+F_0^s}$$

If χ diverges for a finite Z → $F_0^s < 0$
→ attraction (q=0, ω -> 0) between quasiparticles

- in presence of some electron-boson coupling:
  (Ward identity for the density vertex)

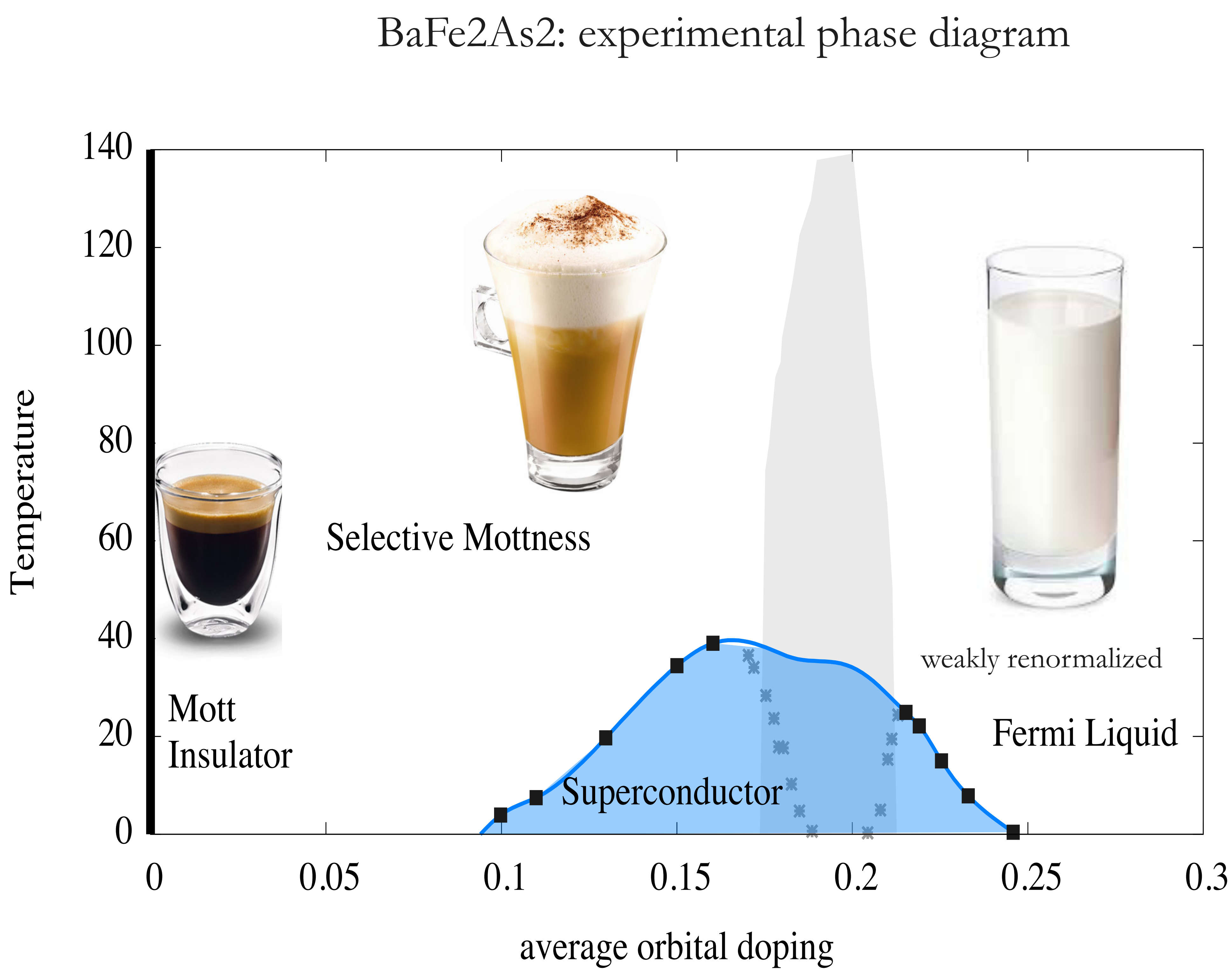
$$\Lambda(q \to 0, \omega = 0) = \frac{1}{Z(1+F_0^s)}$$

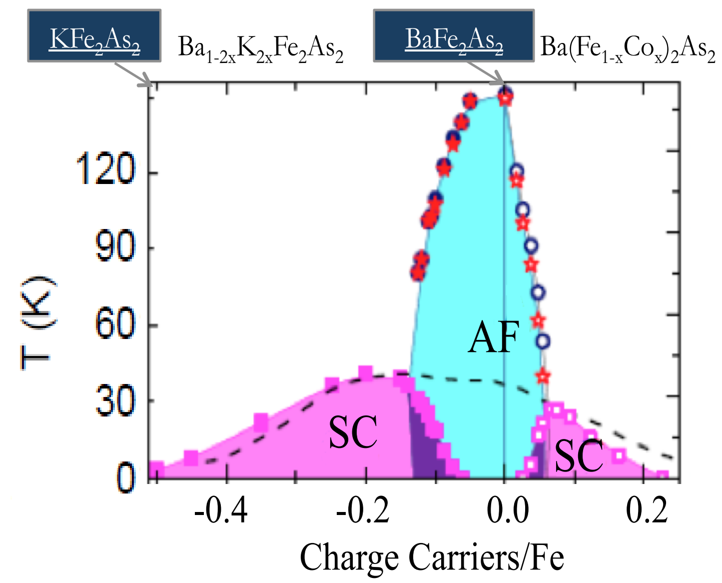
→ enhanced $\sim \chi$

- Phase separation → superconductivity scenario very much studied in the 90's for Cuprates
  cfr: Emery, Kivelson and Lin, PRL 64, 475 (1990)
  Grilli et al. PRL 67, 259 (1991)
  Castellani, Di Castro and Grilli, PRL 75, 4650 (1995) , …

In this region not only the quasiparticle energies are renormalized non-trivially, but also their interactions (mutual and with low-energy bosons)!

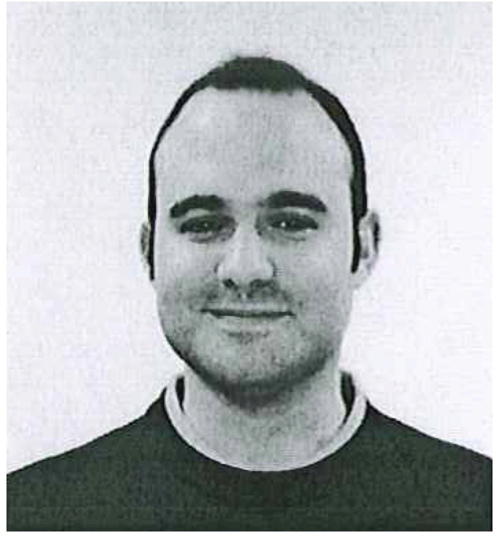
“Best” correlations: at the crossover between weak and selective





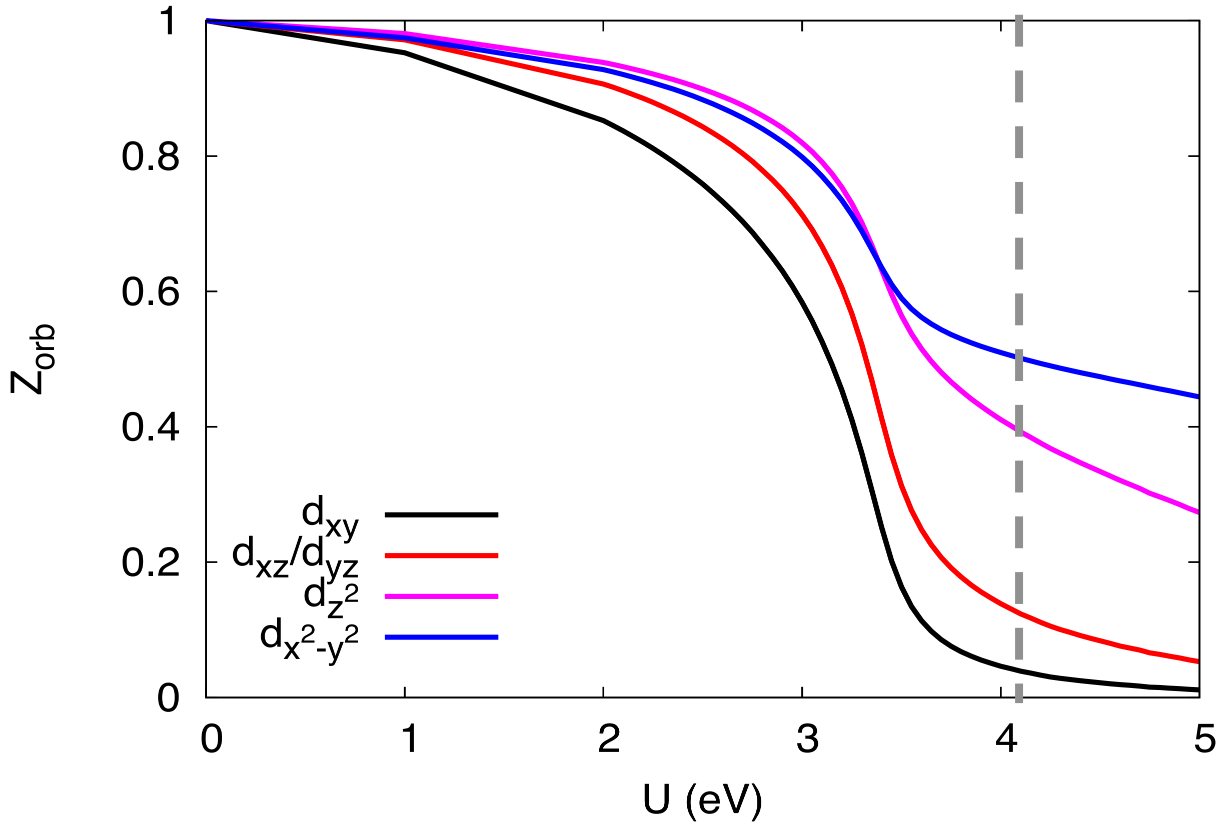
LdM, Giovannetti, Capone, PRL 2014 “Selective Mott Physics as a Key to Iron Superconductors”

FeSe within DFT+Slave-Spin mean-field

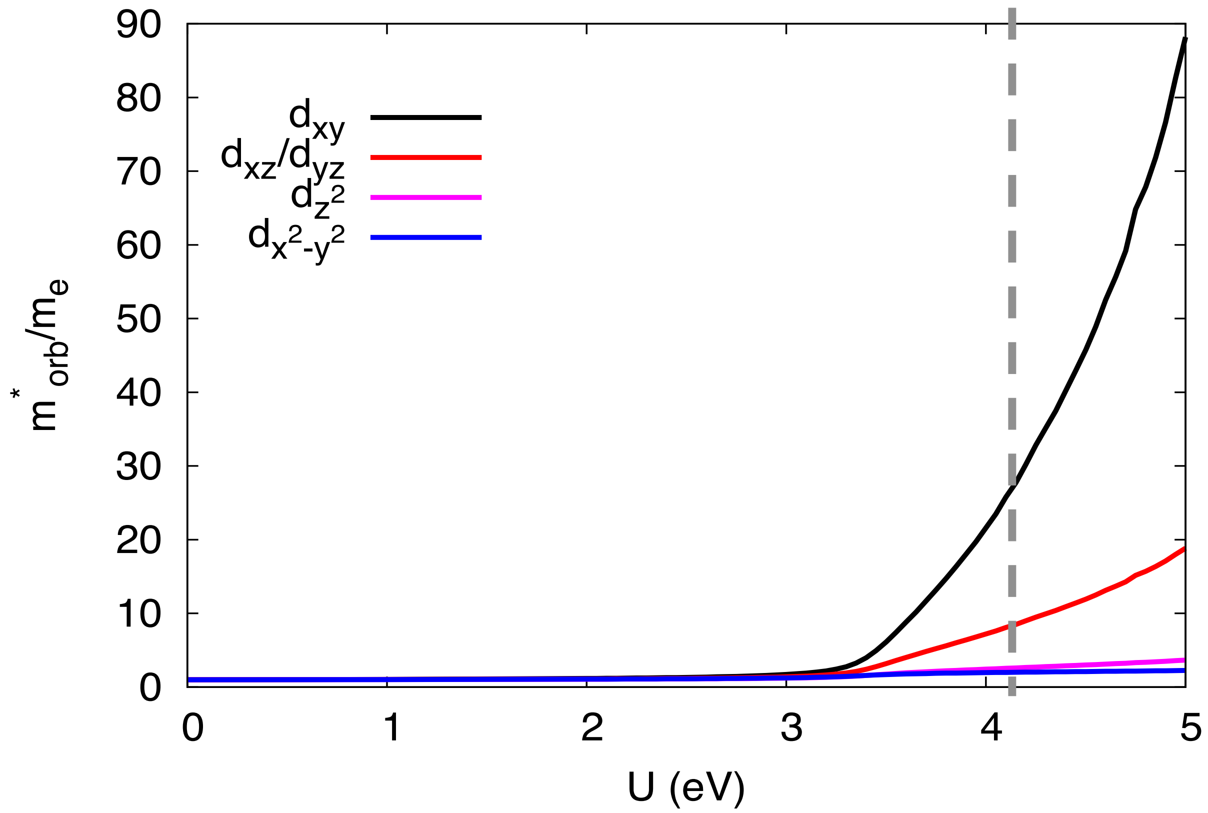


P. Villar Arribi
ESRF

quasiparticle weights



mass enhancements

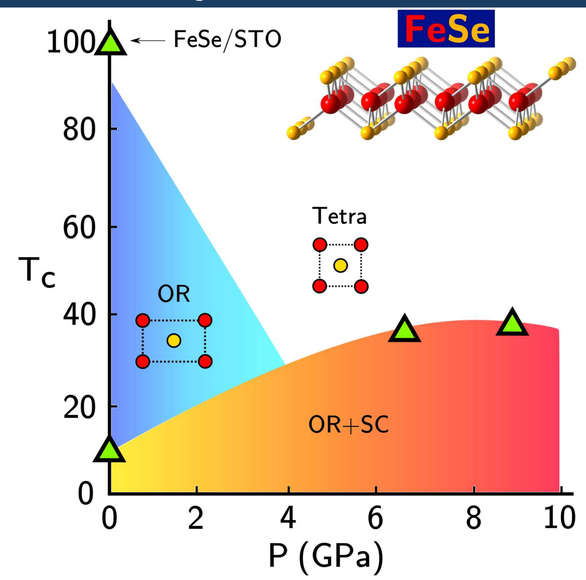


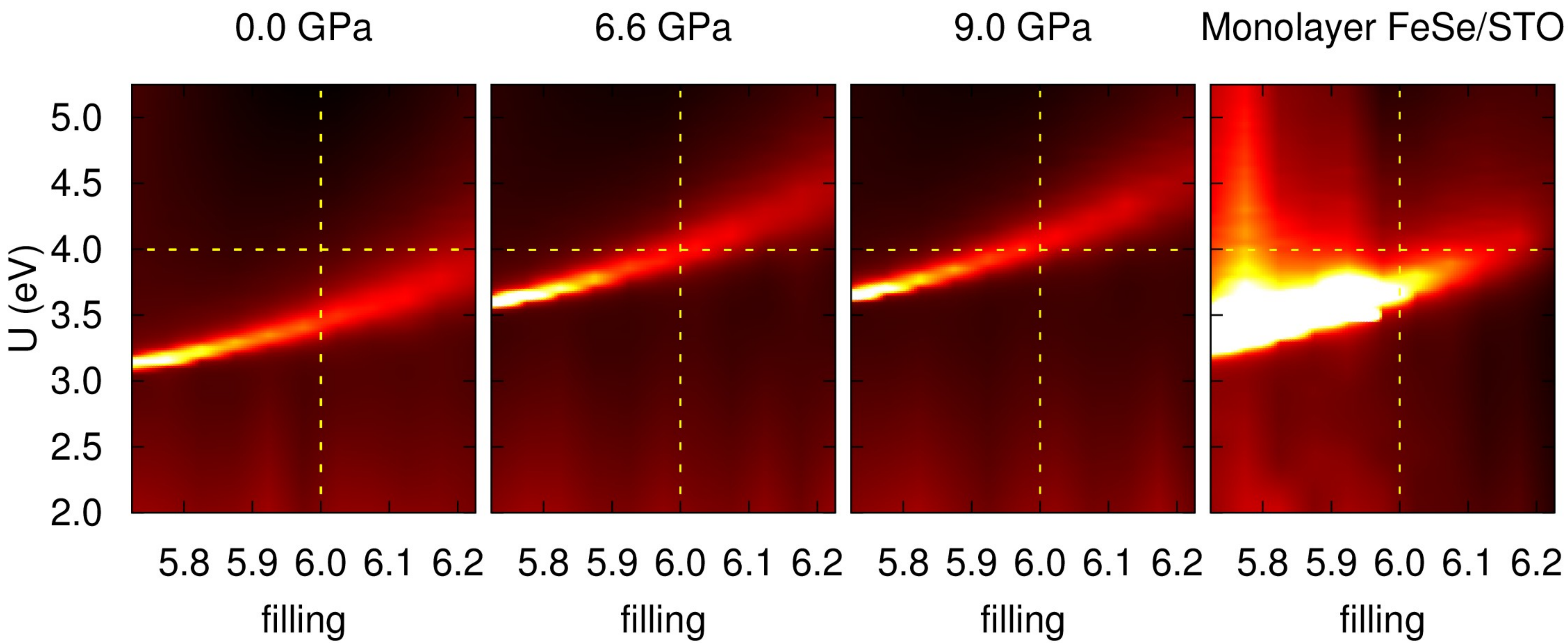
RPA-estimated U : bulk material deep within the "Hund's metal" phase

FeSe/pressure & monolayer

FeSe: Tc grows under pressure

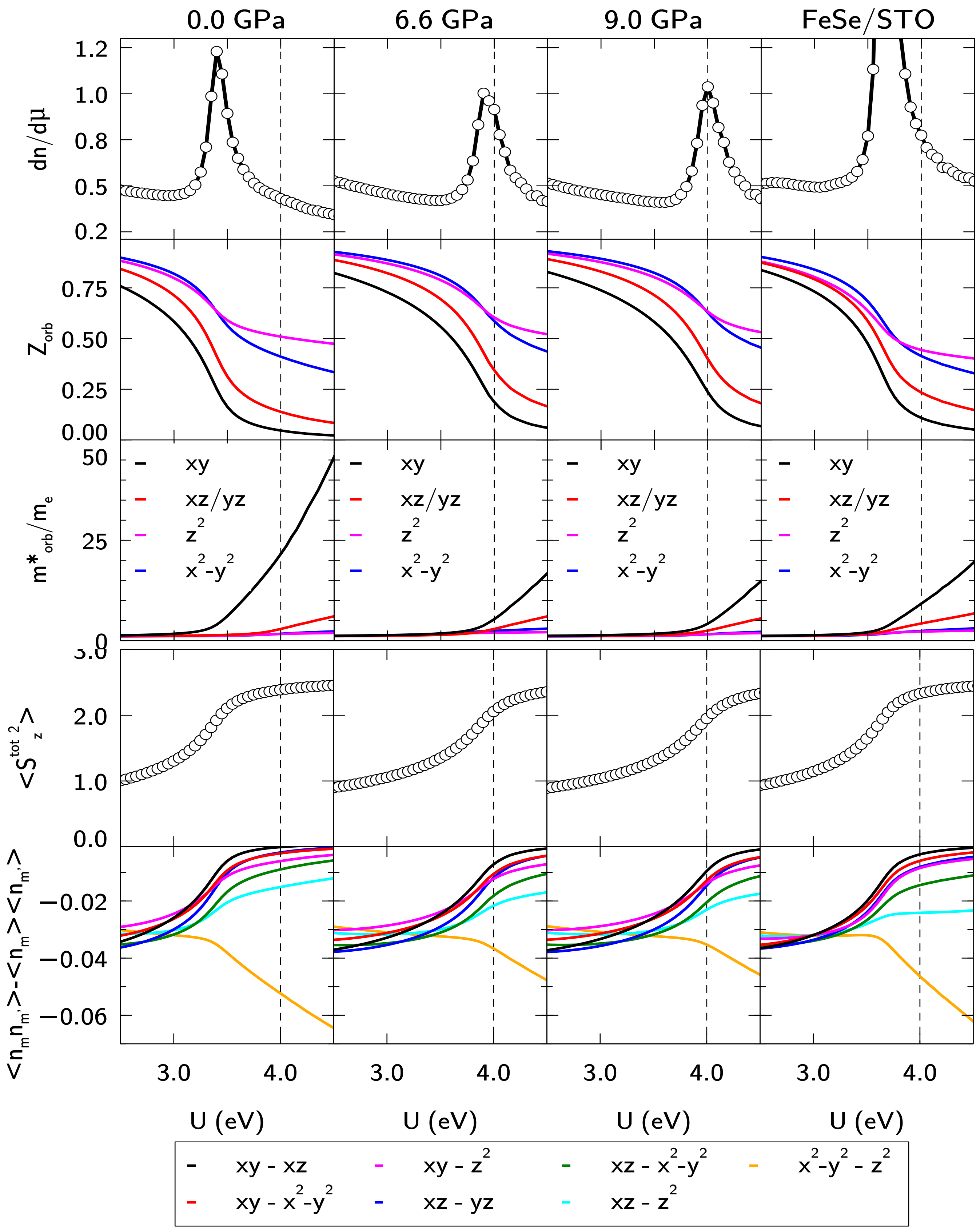
FeSe monolayer: highest claimed T_c

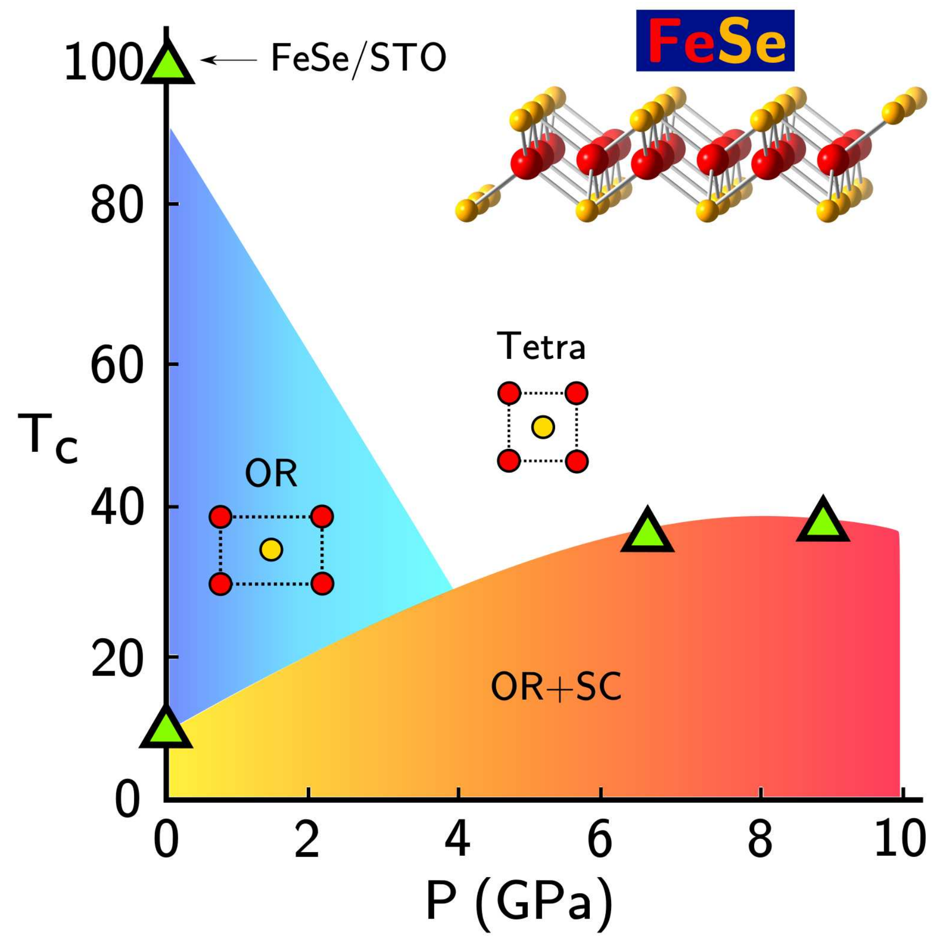




Pablo Villar Arribi and LdM, ArXiv:1803.01494

FeSe/pressure & monolayer





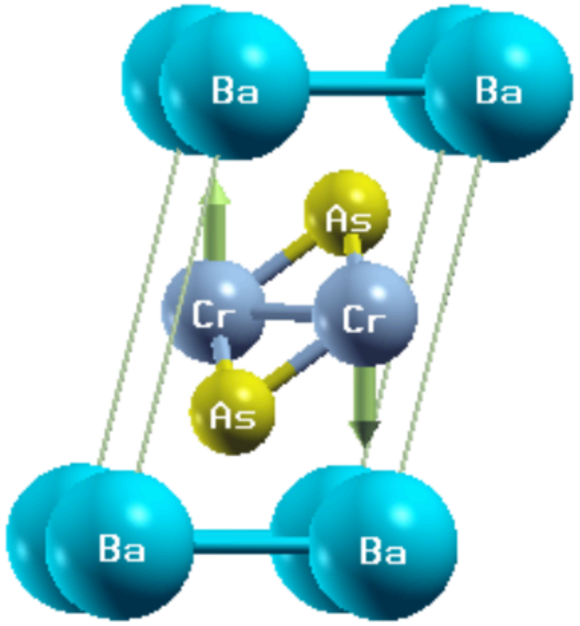
Pablo Villar Arribi and LdM, ArXiv:1803.01494

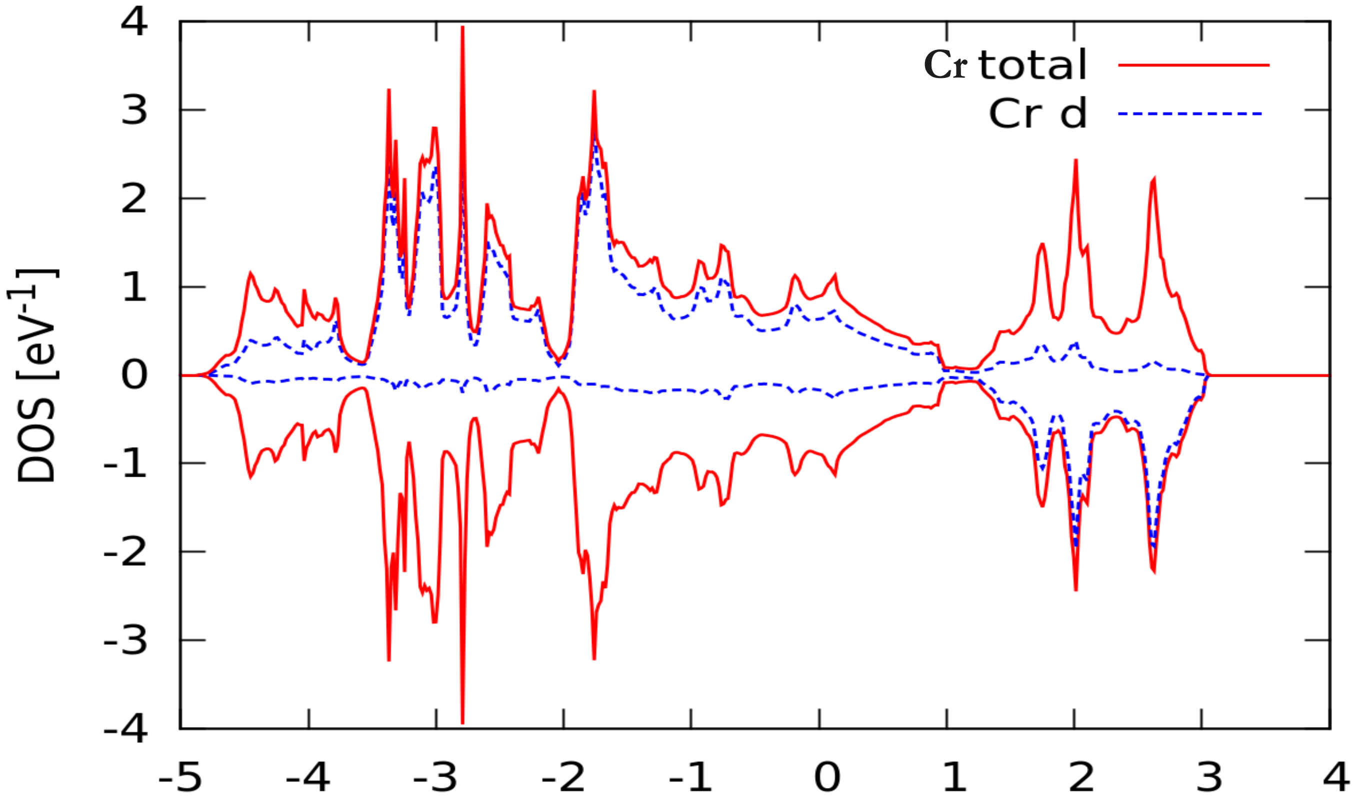
d^4 symmetric of d^6? $BaCr_2As_2$

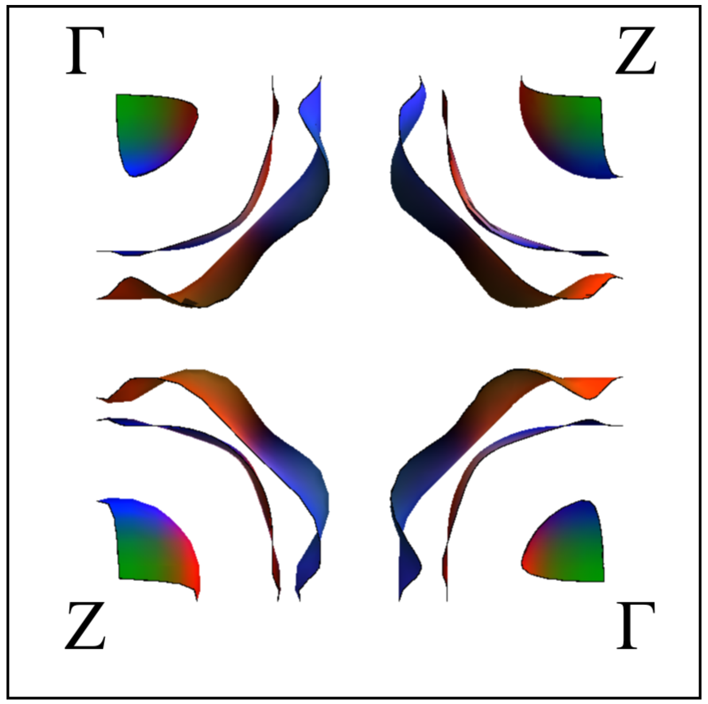
PHYSICAL REVIEW B **95**, 205118 (2017)

Chromium analogs of iron-based superconductors

Martin Edelmann,[1,*] Giorgio Sangiovanni,[1] Massimo Capone,[2,3] and Luca de' Medici[4,5]



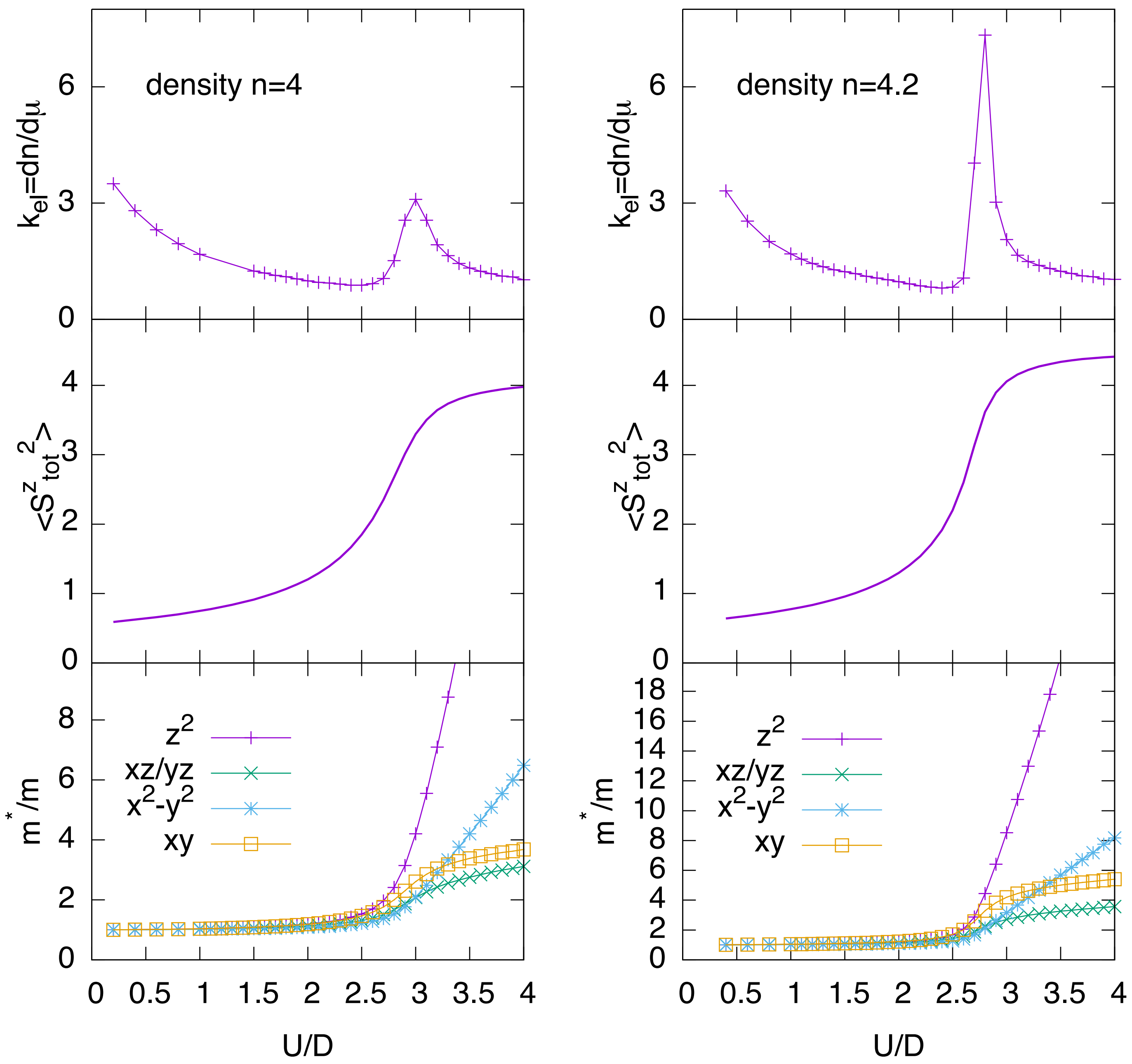




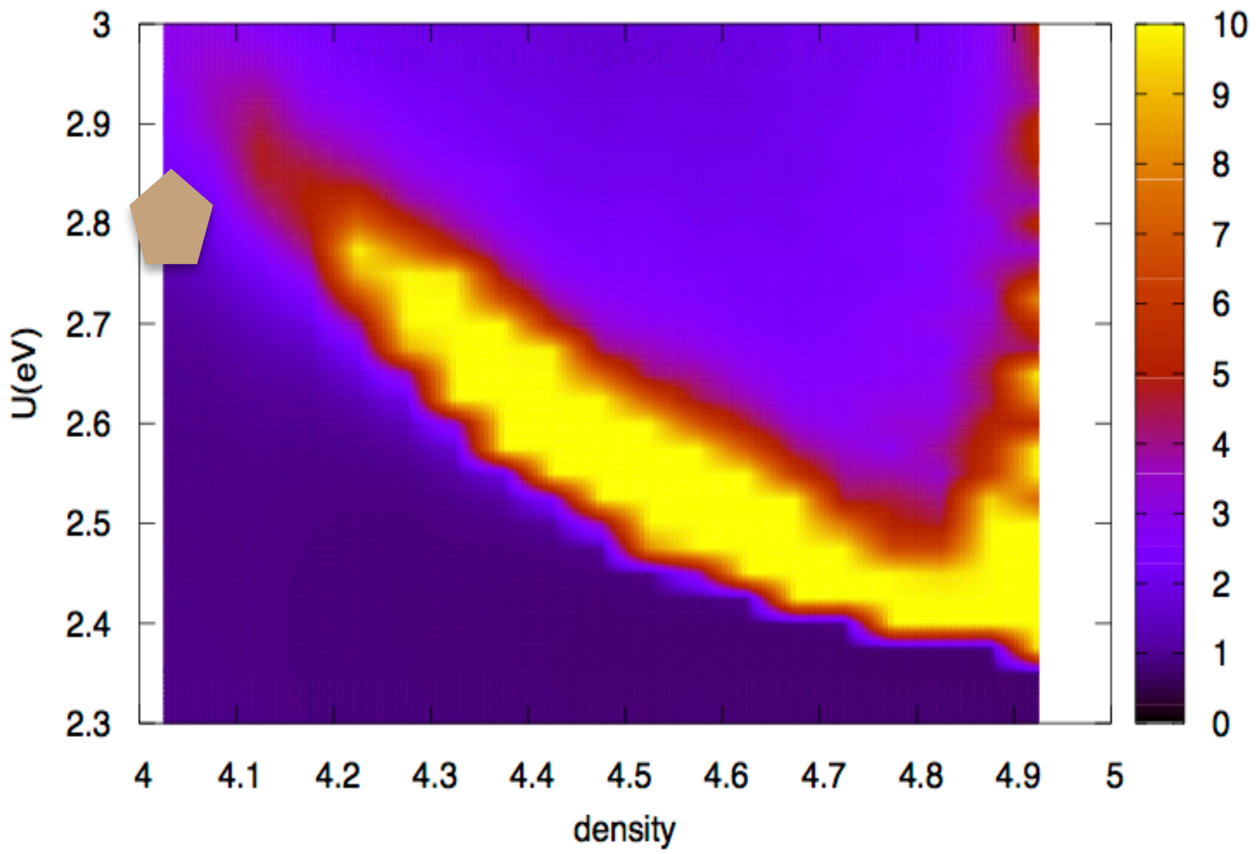
	γ (mJ K^{-2} mol^{-1})
Experiment (from Ref. [32])	19.3
LDA (from Ref. [32])	9.3
GGA (this work)	9.4
GGA+U (this work)	7.2
DFT+DMFT d-d (this work)	13.0
DFT+DMFT Kan (this work)	15.9

$BaCr_2As_2$: Hund's metal phase and compressibility

Paramagnetic phase



Electronic Compressibility

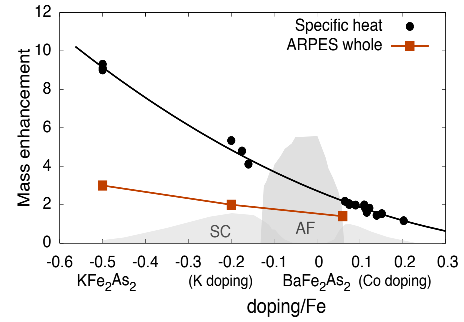


Superconductivity under negative pressure/electron-doping?

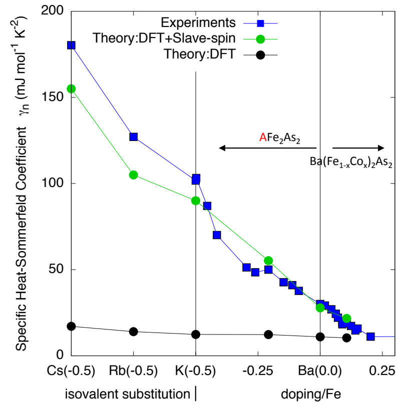
See also: Bascones et al. PRB 95, 075115 (2017)

Conclusions and References

- **Hund's metals: high local moments, enhanced correlations, selective**
- **Onset of orbital selectivity easily highlights the Hund's metal frontier**

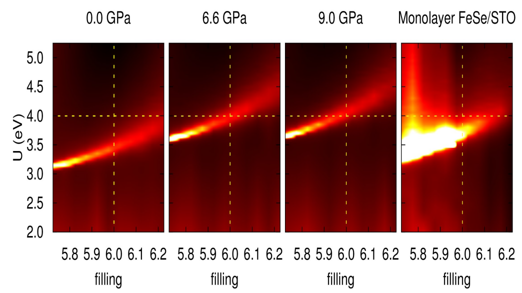


LdM, G. Giovannetti and M. Capone, PRL 112, 177001 (2014)
Selective Mott Physics as a key to Iron superconductors
LdM, *Weak AND strong correlations in Fe Superconductors*,
in "Iron-based Superconductivity", Springer, 211, 409 (2015)
LdM, *Hund's metals, explained*, Juelich lecture, ArXiv:1707.03282



- **Hund's induced phase-separation/enhanced qp interactions at Hund's metal frontier**
- **The mechanism can be tracked by orbital decoupling and could favor superconductivity**

LdM, *Hund's induced Fermi-liquid instabilities and enhanced quasiparticle interactions*
PRL 118, 167003 (2017)



Analogous scenario in FeSe

P. Villar Arribi and LdM, *Hund-Enhanced compressibility in FeSe and its correlation with Tc*,
ArXiv:1803.01494

What about d^4 (Cr-122)?

M. Edelmann et al., *Cromium Analogs of Iron-based Superconductors*, PRB 95, 205118 (2017).